Remembering Pavel: detectors for phase contrast x-ray microscopy

Chris Jacobsen
Associate Division Director, X-ray Science Division, Advanced Photon Source
Professor, Physics & Astronomy, Northwestern University

Argonne NATIONAL LABORATORY | Advanced Photon Source

NORTHWESTERN UNIVERSITY

U.S. DEPARTMENT OF ENERGY

How I interacted with Pavel

- I was at Stony Brook (PhD 1983-1988, postdoc 1989-1990, faculty 1991-2009), and involved in a soft x-ray microscope beamline at NSLS.
- Three of my Stony Brook PhD students worked very closely with Pavel: Michael Feser, Benjamin Hornberger, and Christian Holzner.
- I also had the pleasure of having Pavel Jr. in my PHY 251 Modern Physics class, Spring 2009!

A word on Pavel Jr.

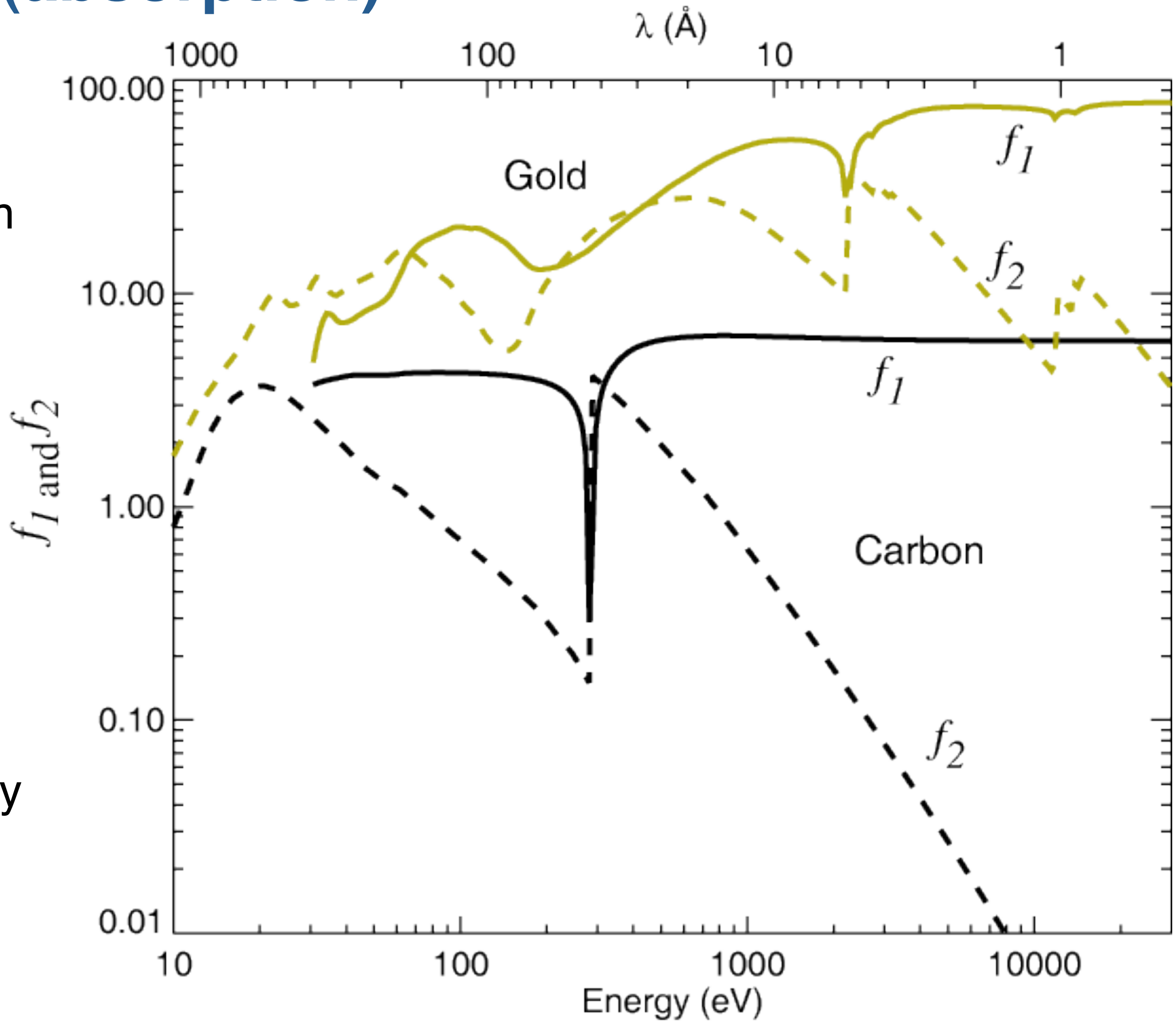
One day in lecture I said something like “an example of quantum interference that’s so obvious even **Stevie Wonder** could see it!”

X ray refractive index: f_1 (phase advance) is stronger than f_2 (absorption)

X-ray refractive index: based on a damped, driven harmonic oscillator model in the limit of high frequency and low damping:

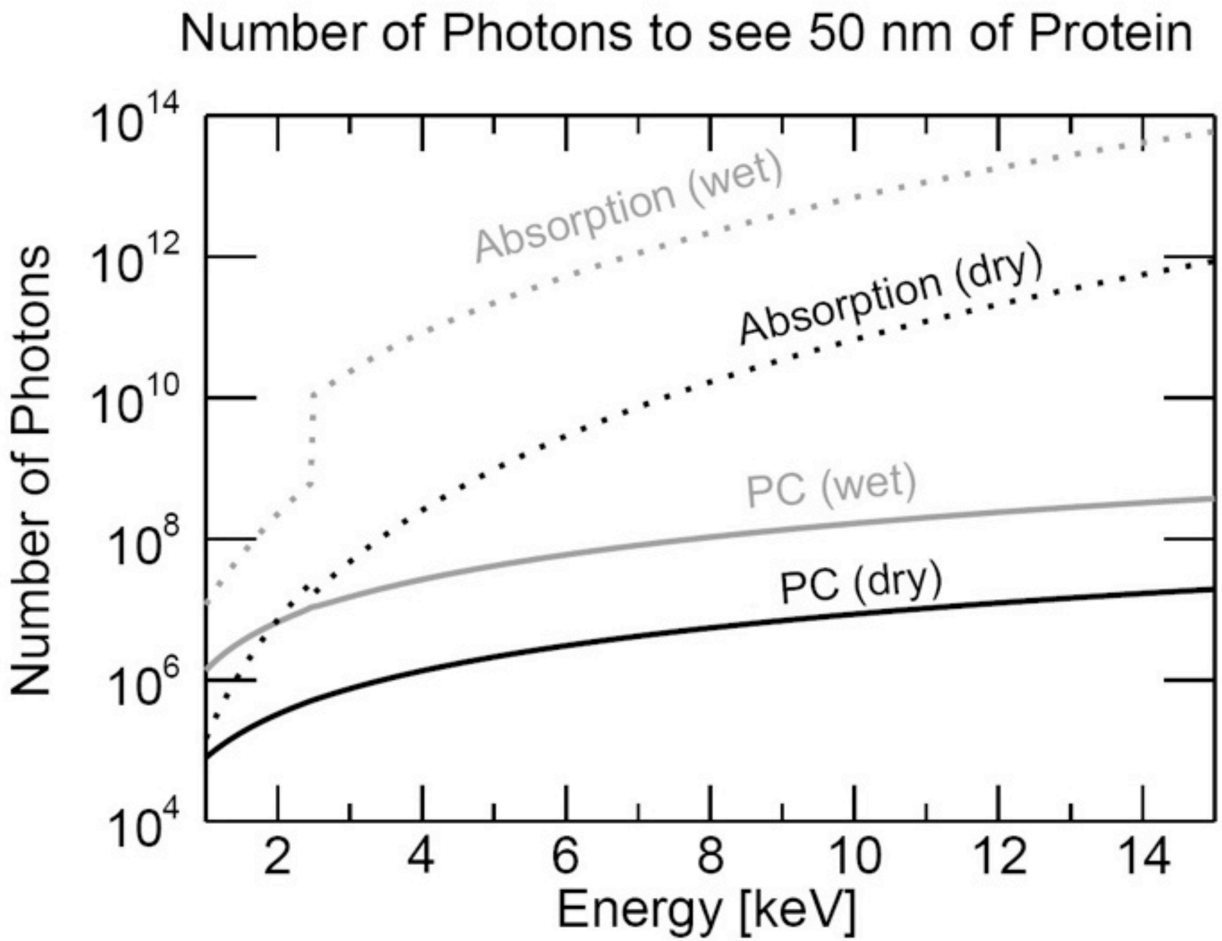
$$n = 1 - \frac{n_a r_e}{2\pi}\lambda^2\left(f_1 + i f_2\right)$$

where n_a gives atoms/volume, r_e=2.82×10^{-15} m is the classical radius of the electron, and (f_1+if_2) specify the atom's oscillator strength at a given photon energy



Phase contrast: soft materials with hard X rays

Phase contrast maximizes contrast and thus minimizes radiation dose and damage



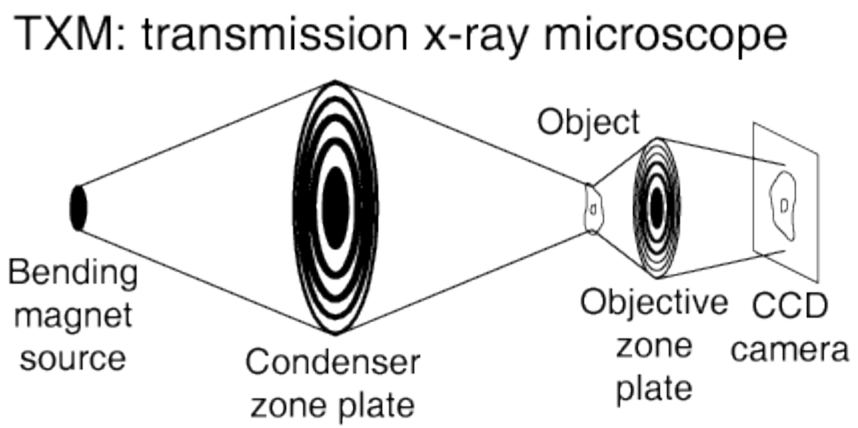
Zone plate microscopes

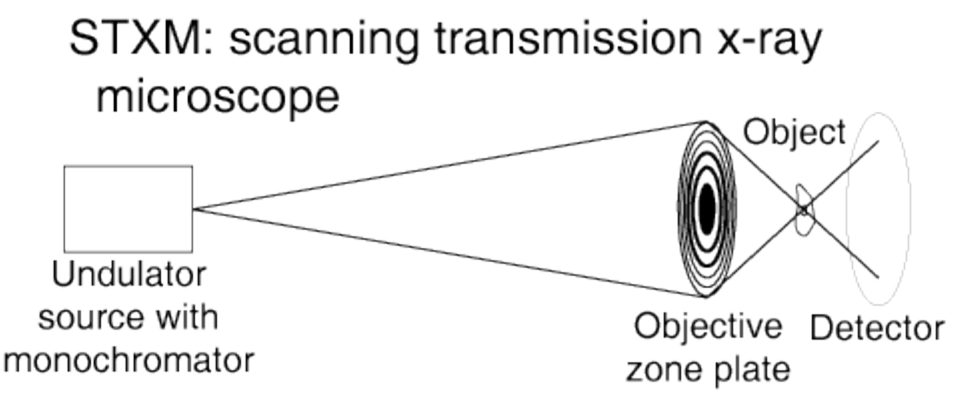
TXM

- Incoherent illumination
- Pixels in parallel: fast
- Higher dose to specimen: ~1-10% optic *after*
- Can work with lab sources

STXM (“sticks-em”)

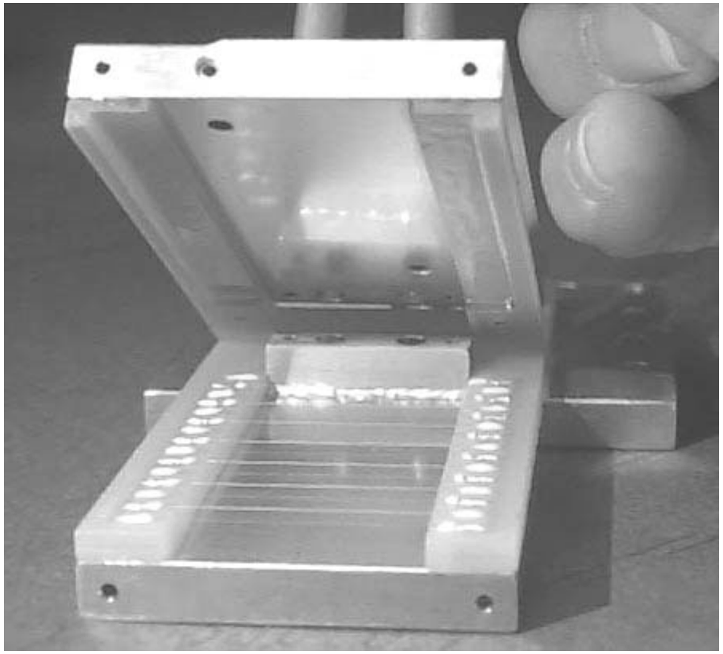
- Coherent illumination
- Pixels in serial: slower
- Lower dose to specimen: ~1-10% efficient optic *before*
- More modalities: fluorescence etc.

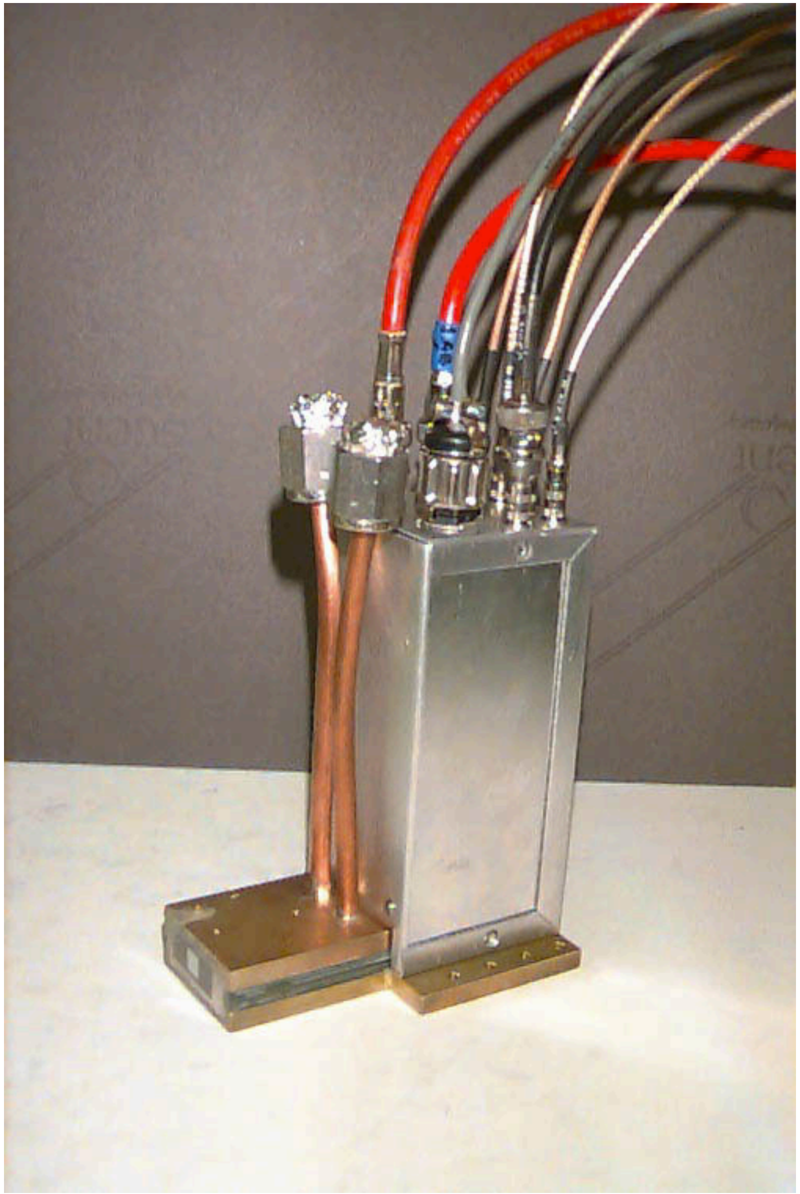




STXM detectors: need for high efficiency

- Gas flow proportional counter at partial atmosphere pressure
- M. Feser, M. Carlucci-Dayton, C. Jacobsen, J. Kirz, U. Neuhäusler, G. Smith, and B. Yu, *Proc. SPIE* **3449**, 19 (1998).





Michael Feser, Graham Smith, Chris Jacobsen

Grasmere (Lake District, England), August 1999

Incorporating phase contrast: Polack and Joyeux

Split focus creates fringes in the far-field; phase gradients produce fringe shifts. Polack and Joyeux, 1993 X-ray Microscopy conference (Chernogolovka, Russia).

We then collaborate with Polack and Joyeux.

Some time later, Pavel visits NSLS X1A with Graham Smith. Let's accept all the light by using a patterned silicon drift detector!

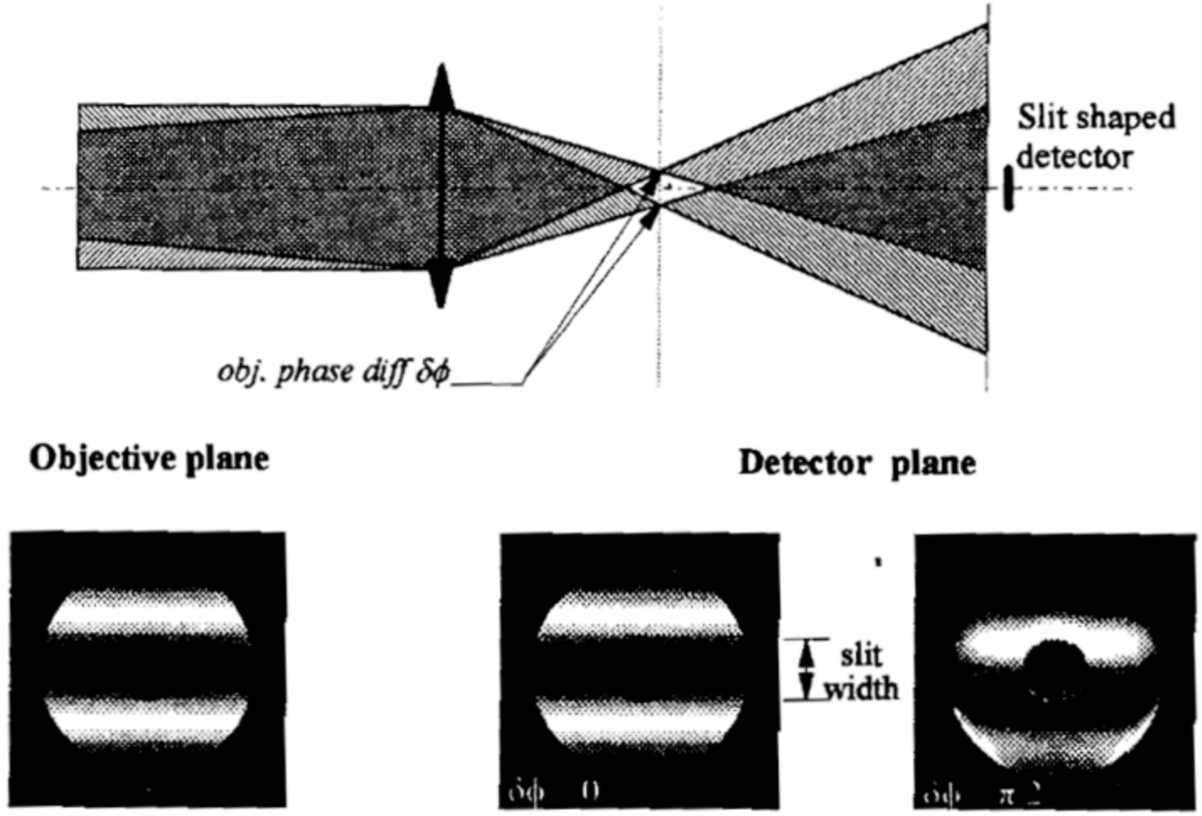
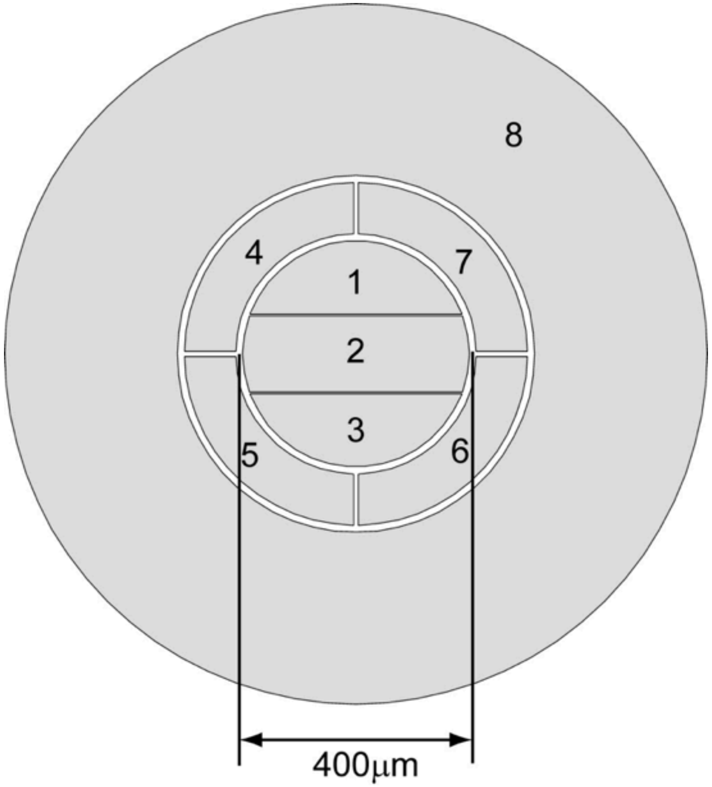


Figure 1. Principle of the detection of a fringe shift

Segmented silicon detector: first generation

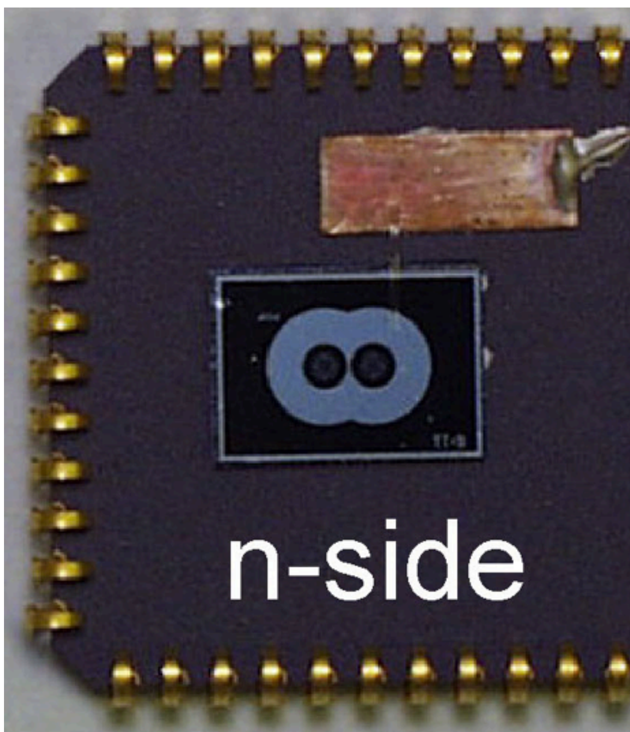
- Pavel dives in!
- Inner region: for interferometric phase contrast
- Middle region: quadrant annular bright field
- Outer region: annular dark field
- Signal levels are challenging:
  - 10^5 photons/sec, 350 eV/photon: 10^7 e-/sec or ~1 pA, with desired pixel time of 1 msec
  - Charge integration followed by analog signal readout

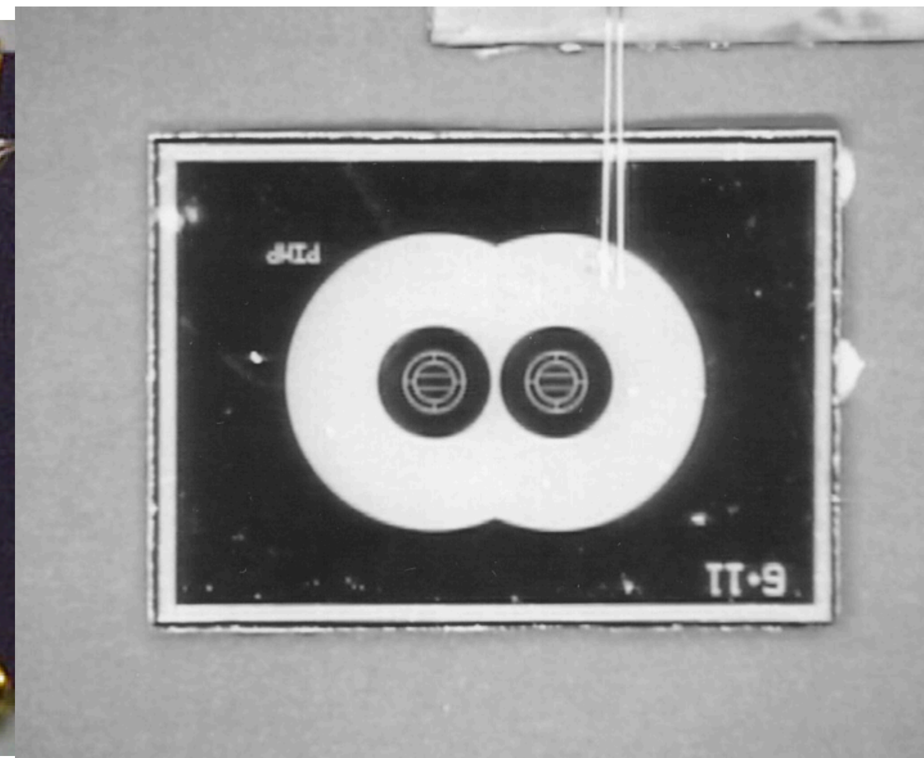


Pavel and friends dive deeper!

- First prototypes fabricated at BNL.
- Production silicon drift diode chips fabricated by Lothar Strüder, Peter Holl, and collaborators at Max-Planck-Institut für extraterrestrische Physik, Passing, Germany (at that time)
- Shallow p-type implant in high resistivity n-type silicon, with 90 Volt bias for n-side illumination.

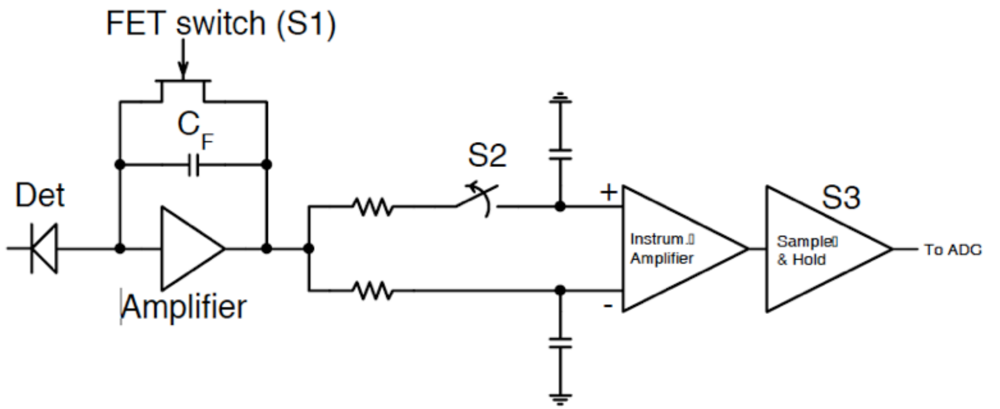
Twinned active regions for subtraction of microphonics

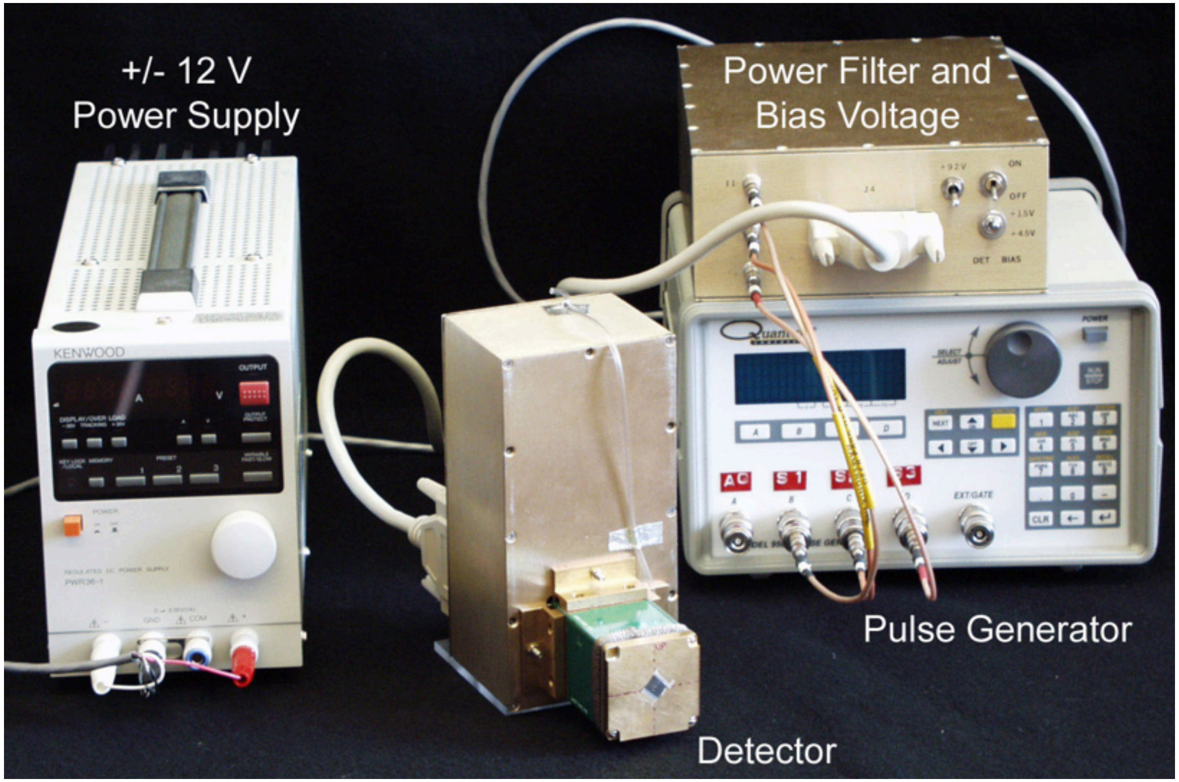




Charge-integrating electronics for low-flux soft X rays

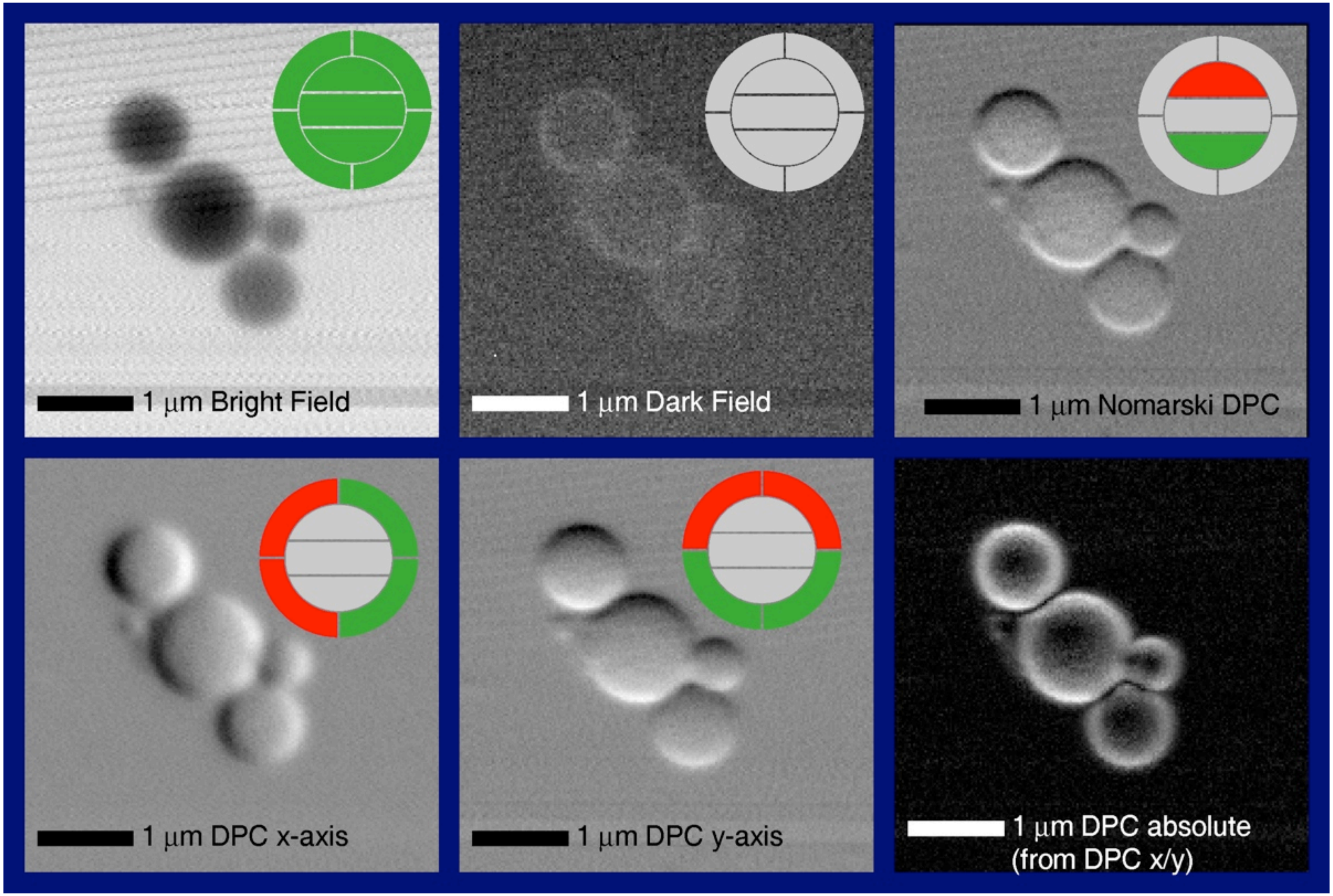
M. Feser, C. Jacobsen, P. Rehak, G. DeGeronimo, P. Holl, and L. Strüder, *Proc. SPIE* **4499**, 117 (2001)





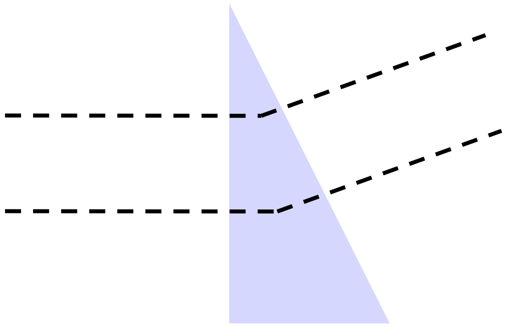
Imaging silica spheres at 520 eV

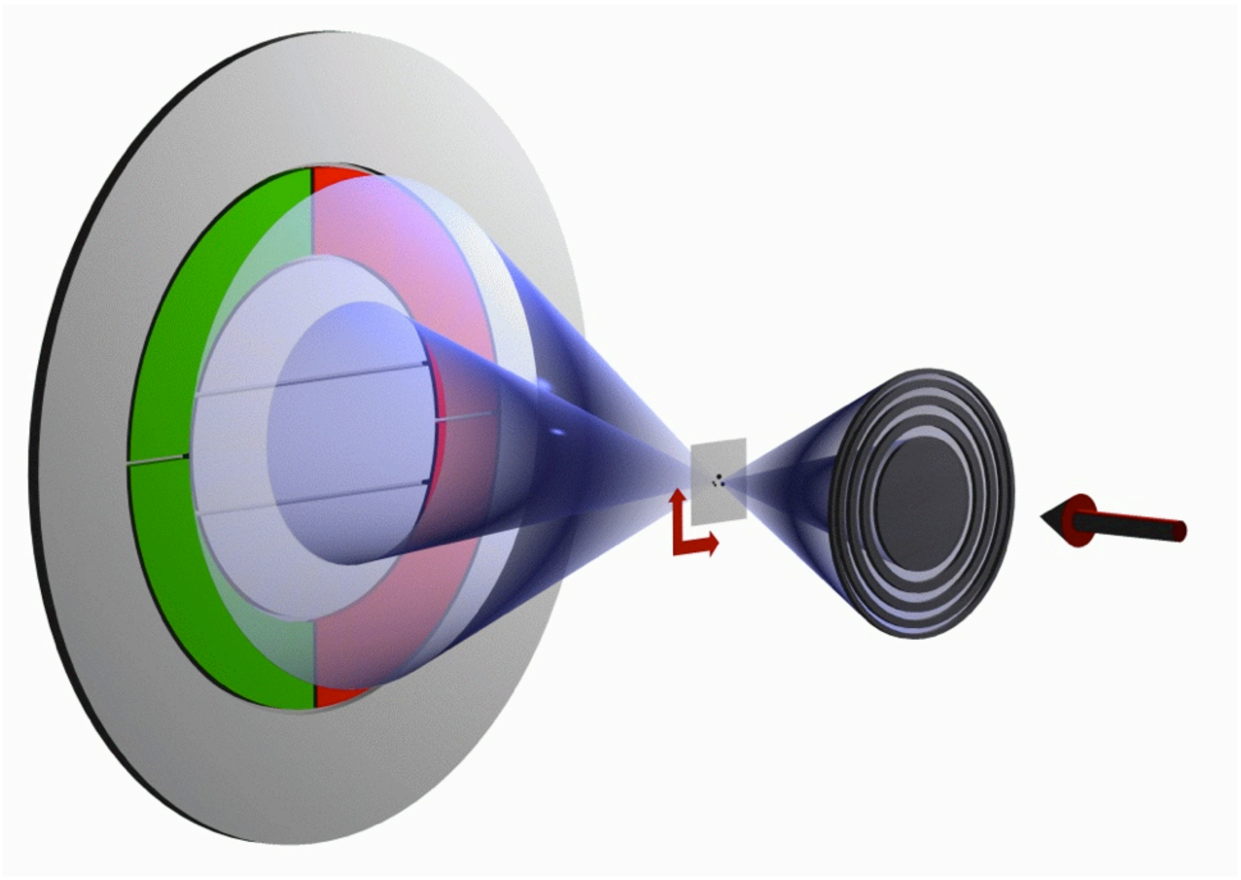
Nomarski works well - but so does simple differential phase contrast!



Differential Phase Contrast

- Refraction model – effect of **phase gradient** (like prism for visible light) :





X-ray Microscopy 1999 conference

Young man, there's no need to feel down
I said young man, get your PhD off the
ground
I said young man, 'cause you're at the
Long Island Sound
There's no need to be unhappy

It's fun to work at the NSLS
It's fun to work at the NSLS
They have everything for
you men to enjoy
You can play with the
vacuum toys

Michael Feser on working with Pavel

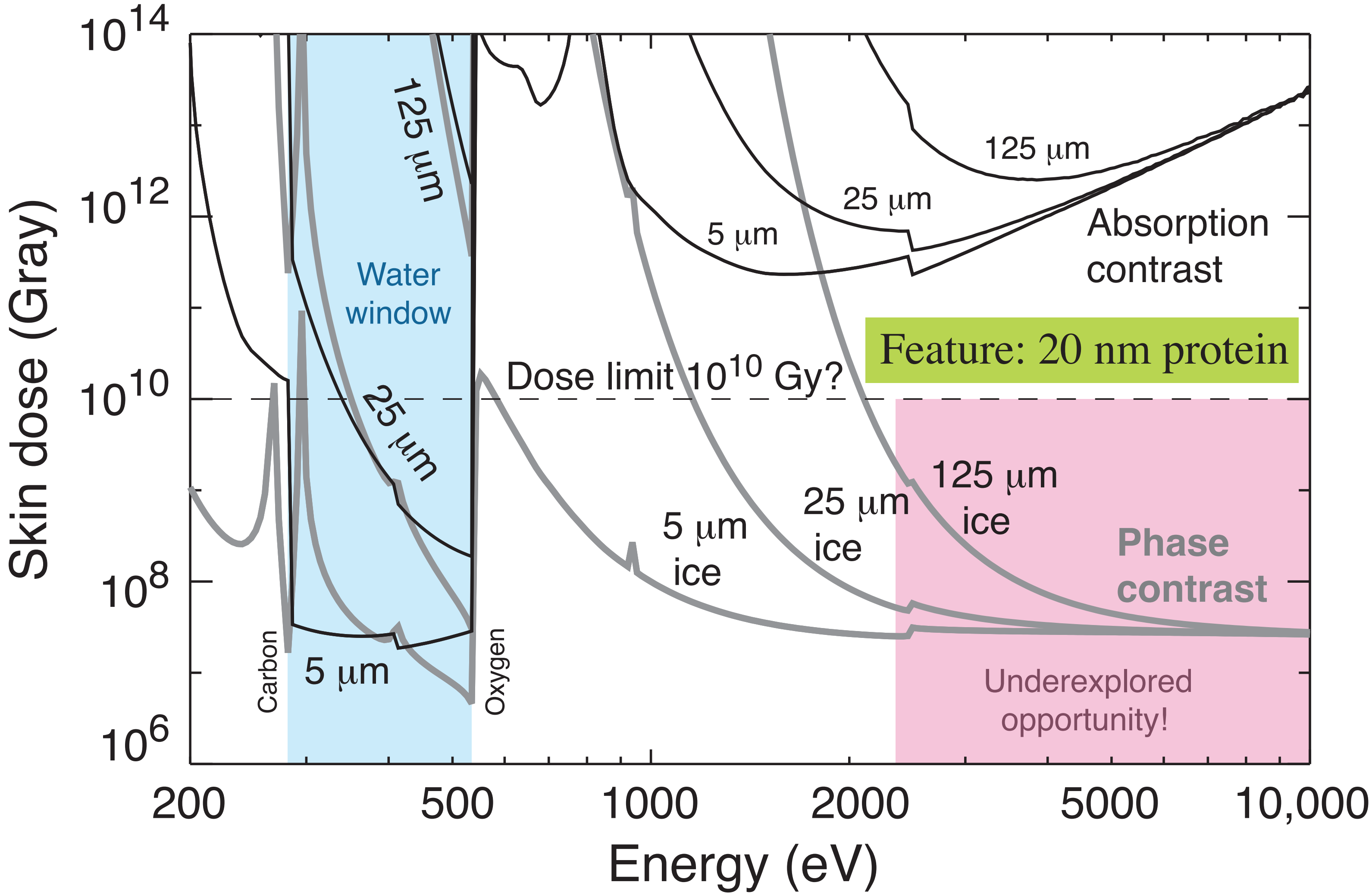
PhD, May 2002: “Scanning Transmission X-ray Microscopy with a Segmented Detector”

Handoff: Michael Feser to Benjamin Hornberger

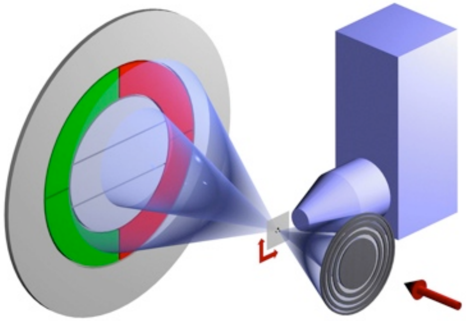
Benjamin Hornberger, PhD, 2007: "Phase Contrast Microscopy with Soft and Hard X-rays Using a Segmented Detector"

From soft to hard X rays

Phase contrast is more important with hard X rays

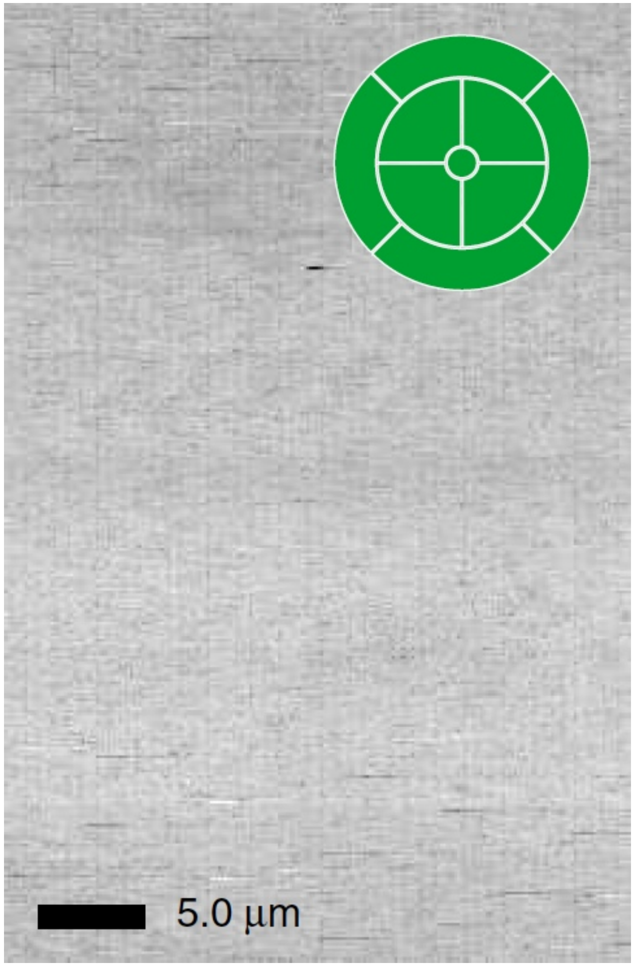


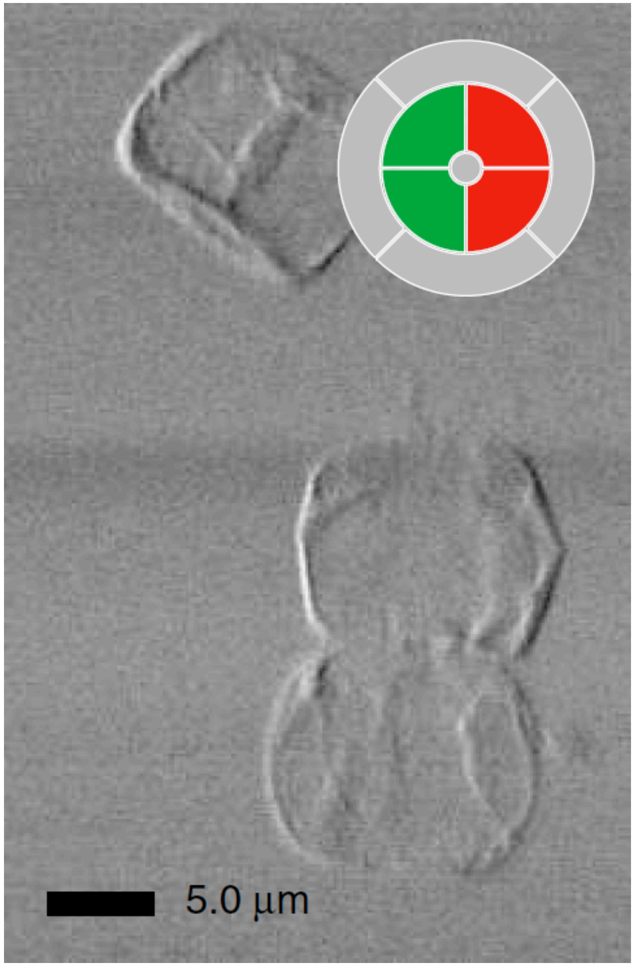
Hard X ray imaging of soft materials



Diatoms at 10 keV (with Stephen Baines and Ben Twining, Stony Brook Marine Sciences Research Center; and Stefan Vogt, Advanced Photon Source at Argonne)

Absorption at 10 keV

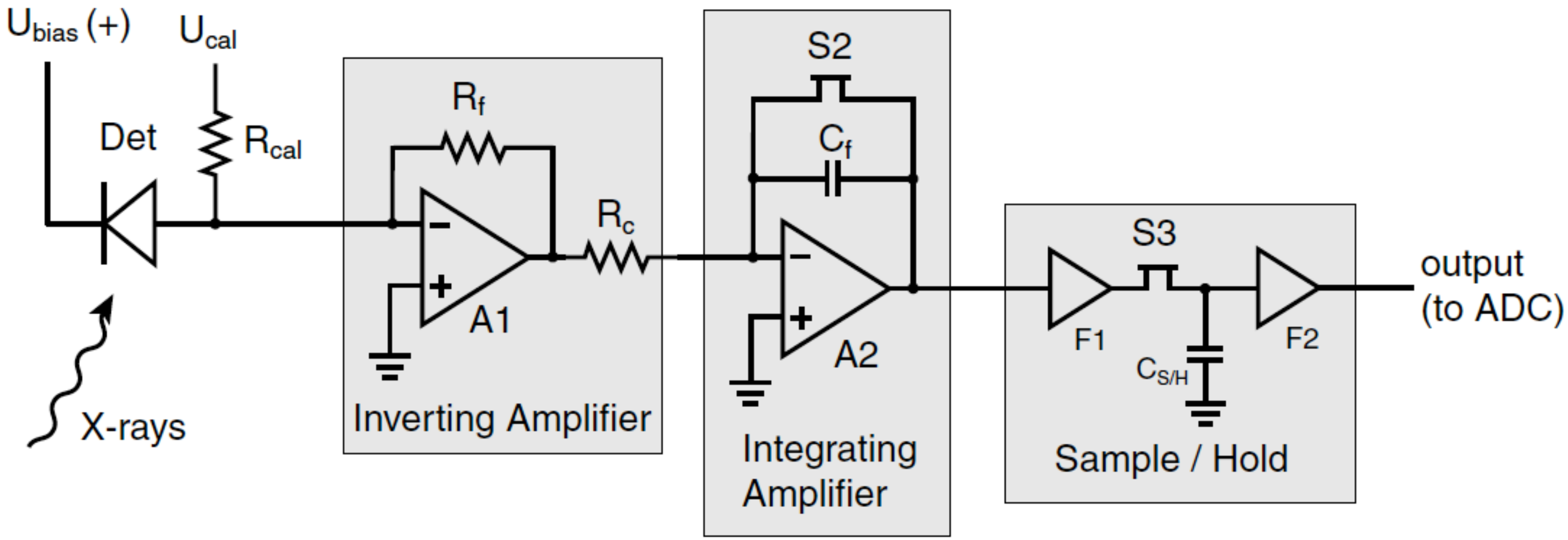




Differential phase contrast

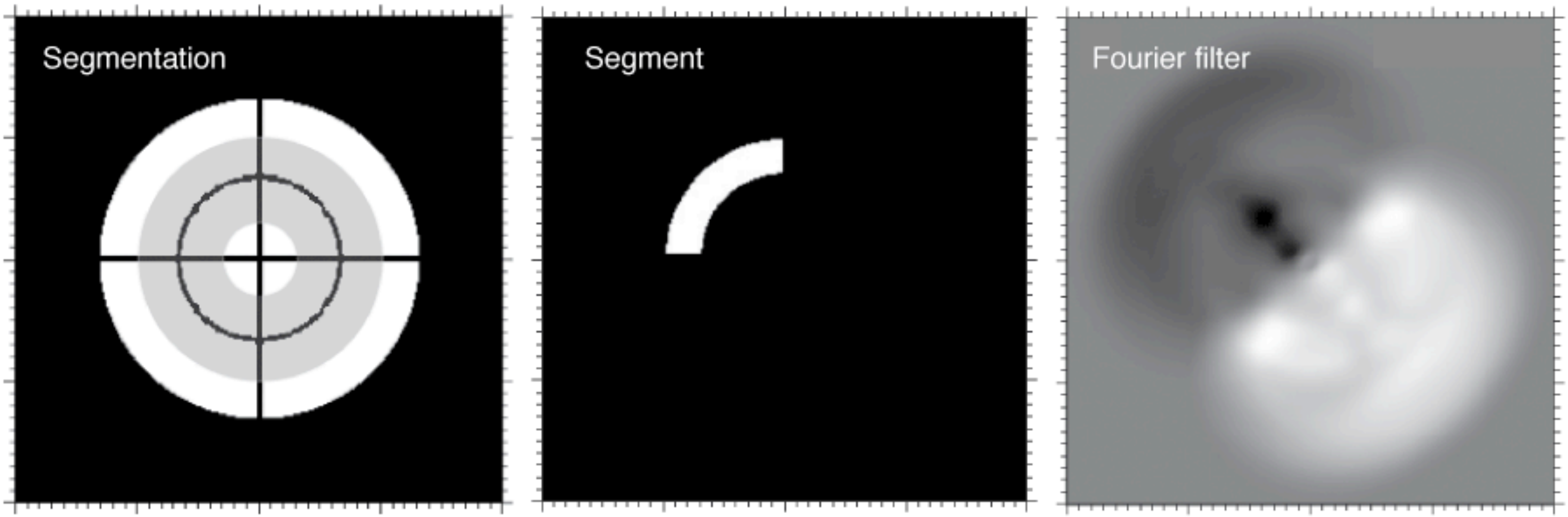
Harder is easier!

- Flux goes from $10^{5\text{-}6}$ to $10^{8\text{-}10}$ photons/sec, and photon energy goes from 500 eV to 10,000 eV: current goes from pA to µA.
- First tests were done with the soft x-ray detector and several layers of aluminum absorber cut from a Coke can!
- B. Hornberger, M.D. de Jonge, M. Feser, P. Holl, C. Holzner, C. Jacobsen, D. Legnini, D. Paterson, P. Rehak, L. Strüder, and S. Vogt, *J. Synchrotron Radiation* **15**, 355 (2008).



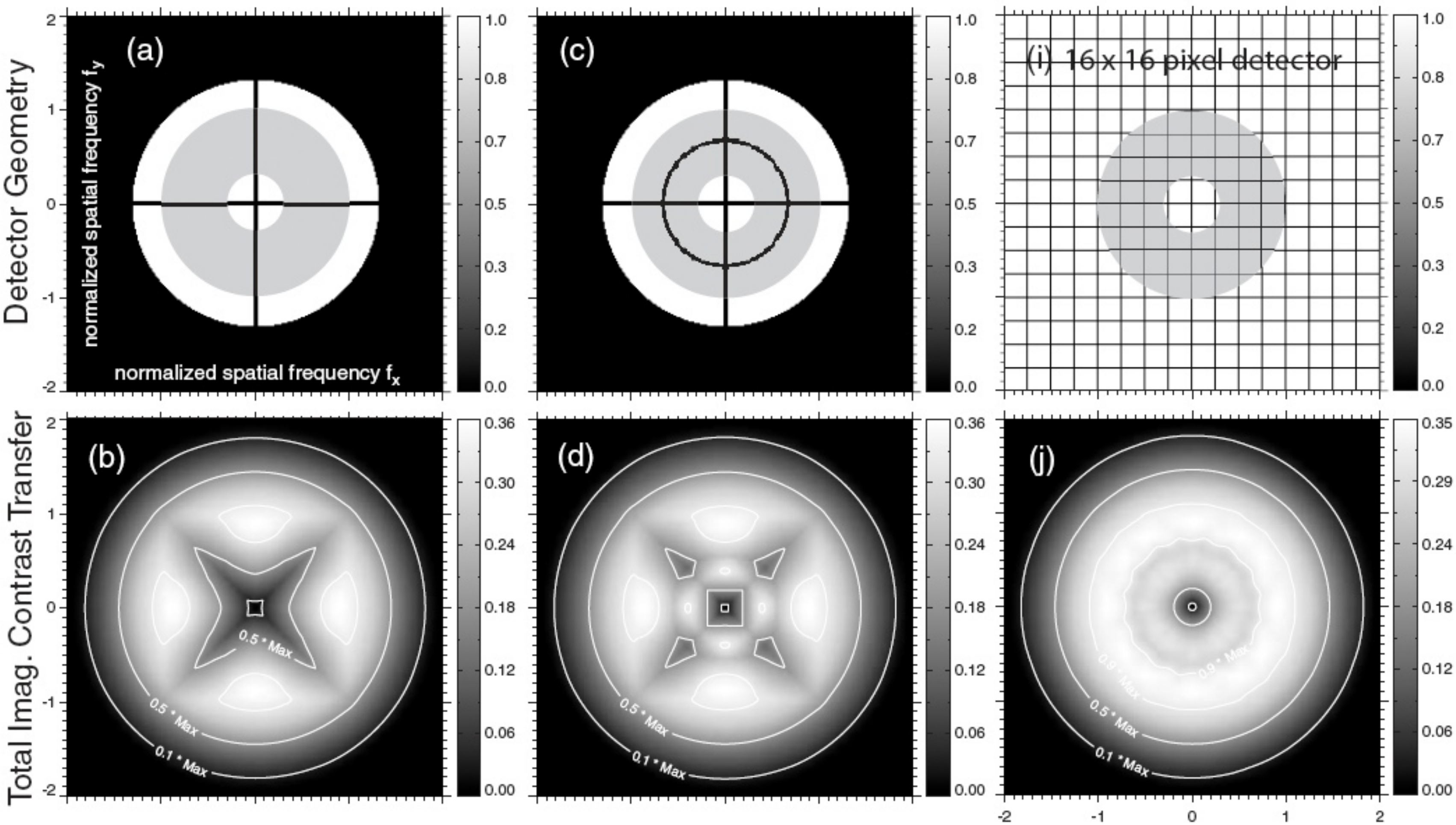
Segmented detector and Fourier filter reconstruction

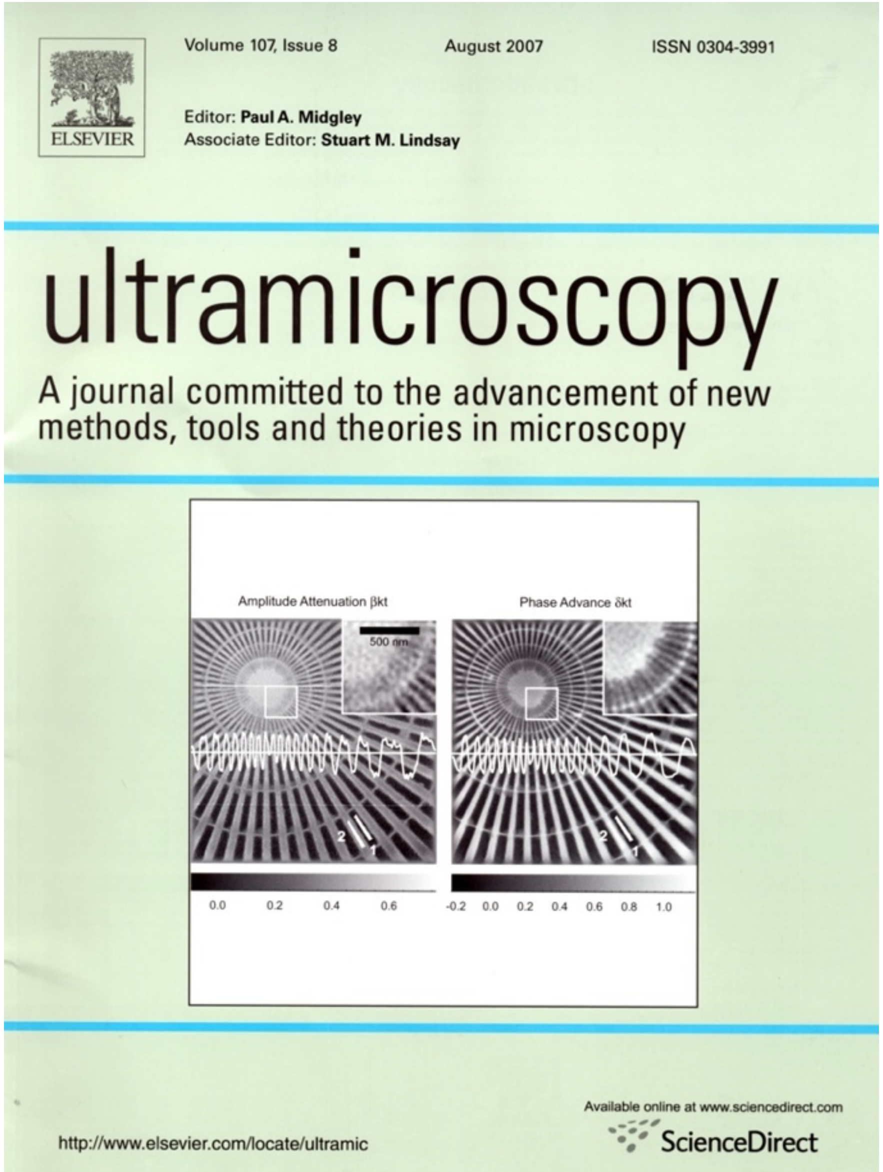
- Limited number of segments means fast readout in scanning microprobe, and fast reconstruction.
- Fourier filtering approach: inspired by STEM work of McCallum, Landauer, and Rodenburg, *Optik* **103**, 131 (1996).
- Initiated by Michael Feser, and made quantitative by Benjamin Hornberger.



Fewer segments=fast readout

Fourier plane coverage of various detector schemes (B. Hornberger PhD dissertation, 2007)





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Specimen
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Zone plate
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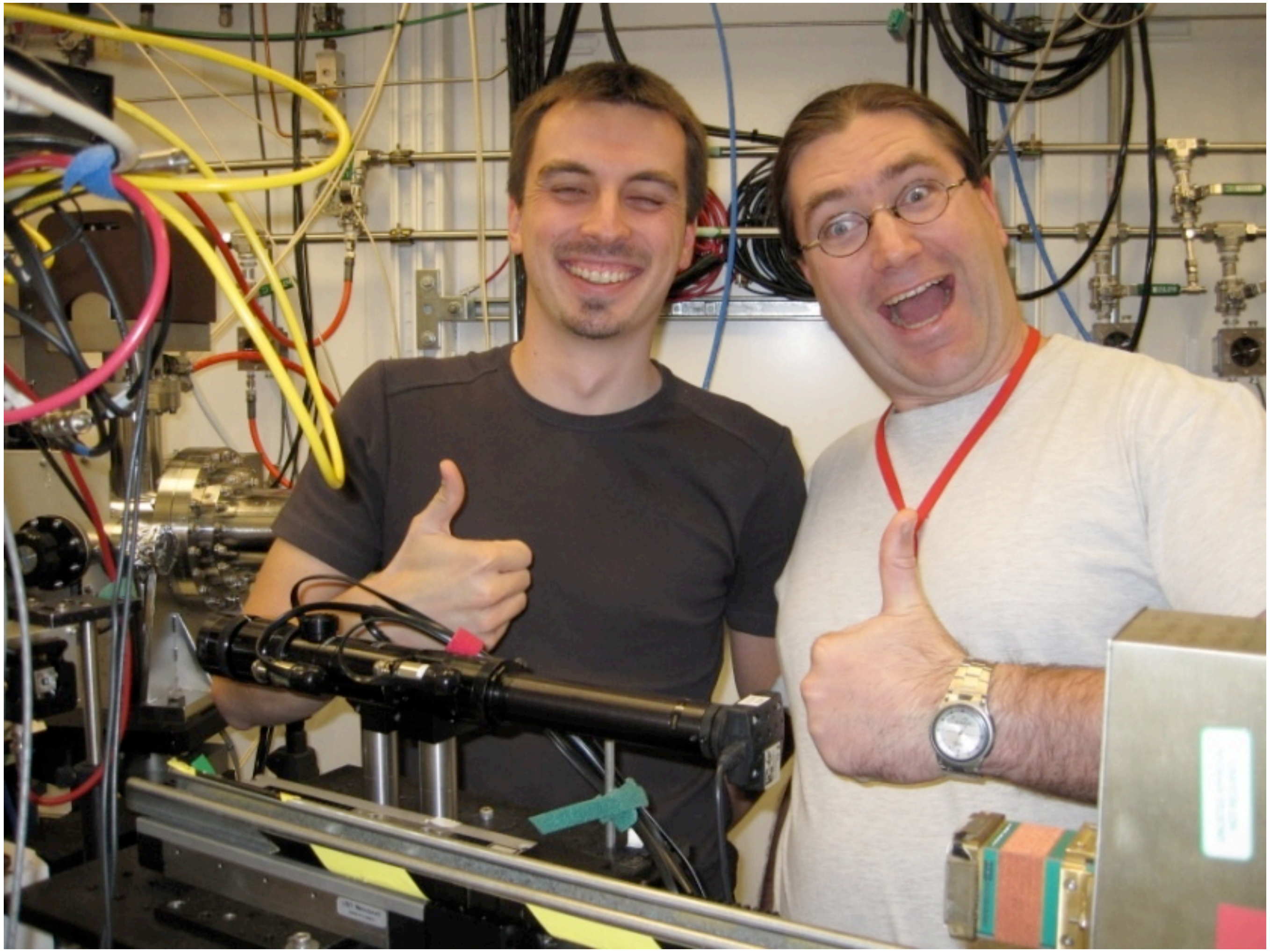
Benjamin Hornberger on working with Pavel

PhD, 2007: “Phase Contrast Microscopy with Soft and Hard X-rays Using a Segmented Detector”

Next in the chain: Christian Holzner

PhD, August 2010: “Hard x-ray phase contrast microscopy - techniques and applications”



Three methods for quantitative phase contrast

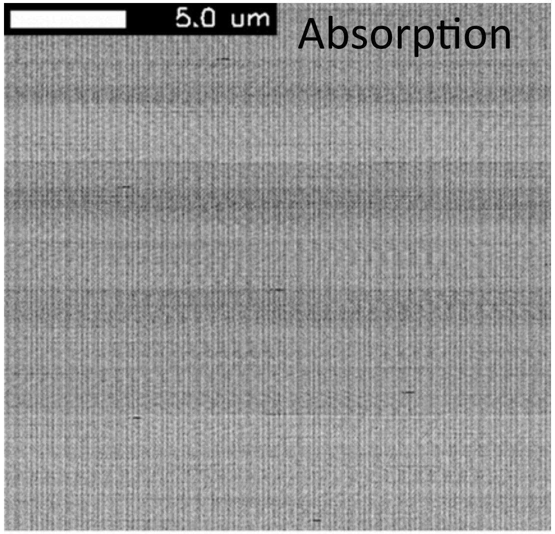
- Fourier filtering method: further theoretical development, improved absolute quantitation
- Fourier integration method from differential phase $\phi = \delta k t$: simple implementation, better at low spatial frequencies

$$(\delta k t) = \mathcal{F}^{-1}\left[\frac{\mathcal{F}[\nabla_x(\delta k t) + i\nabla_y(\delta k t)]}{2\pi i(u + iv)}\right]$$

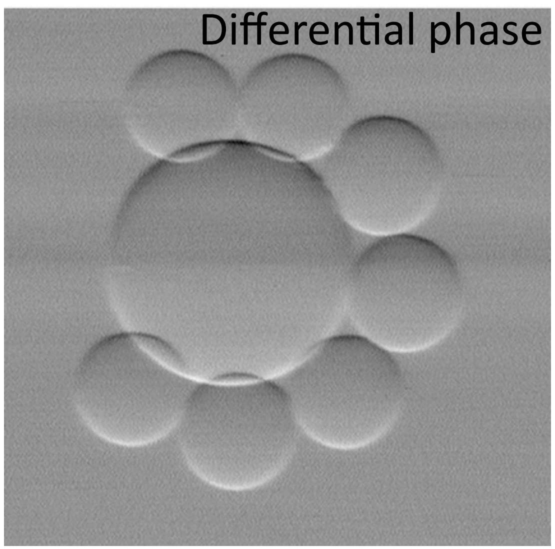
  – M. D. de Jonge, B. Hornberger, C. Holzner, D. Legnini, D. Paterson, I. McNulty, C. Jacobsen, and S. Vogt, *Phys. Rev. Lett.* **100**, 163902 (2008)

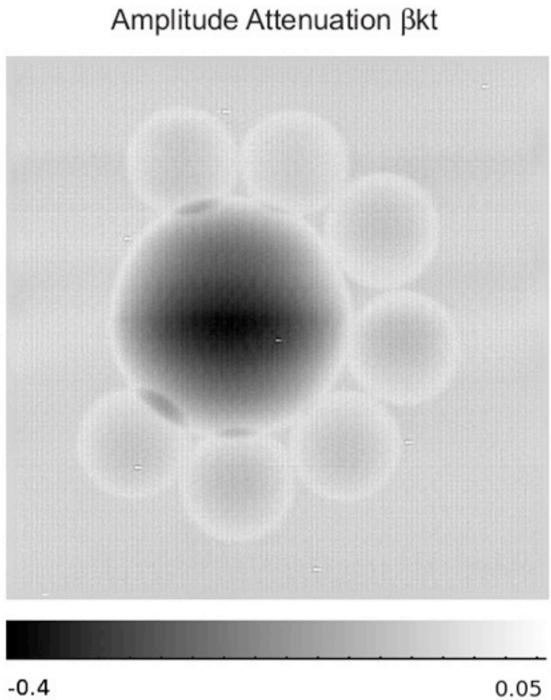
- Zernike phase contrast using the annular detector segments: direct, online images
  – C. Holzner, M. Feser, B. Hornberger, S. Vogt, C. Jacobsen, *Nature Physics* (online: doi:10.1038/nphys1765)

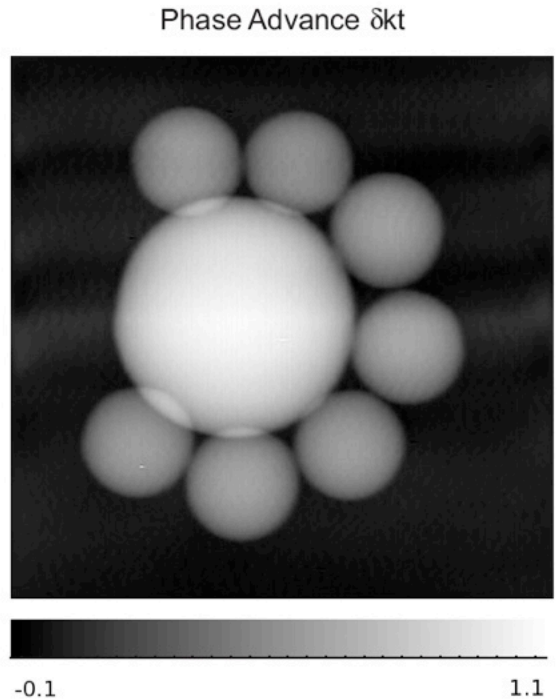
Polystyrene spheres at 10 keV: quantitative



- 50 nm steps, 10 msec pixel clock
- Reconstructed values agree within 2% of Henke tabulation
- Sensitivity: $\pi/180$
- C. Holzner *et al.*

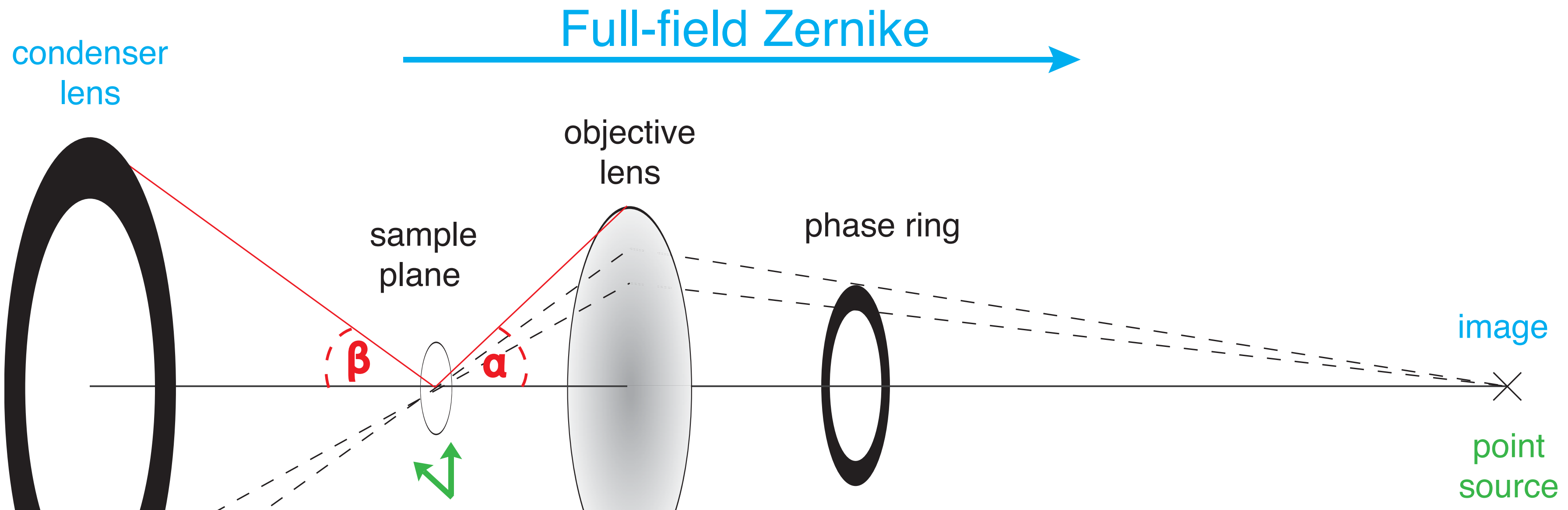


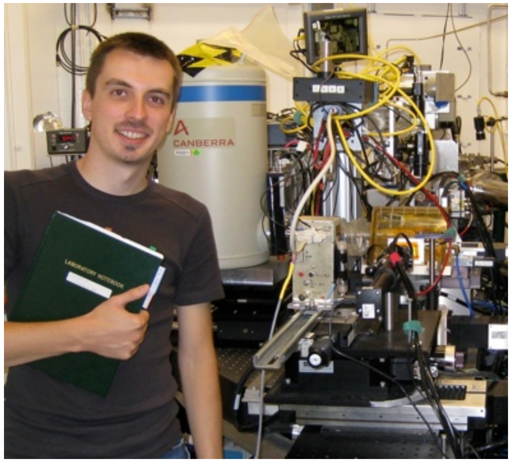




Zernike in a scanning x-ray microscope!

Reciprocity. Suggested by Wilson & Sheppard (1984); US Patent 4,953,188, Siegel, Schmahl, and Rudolph (1990); but not realized.

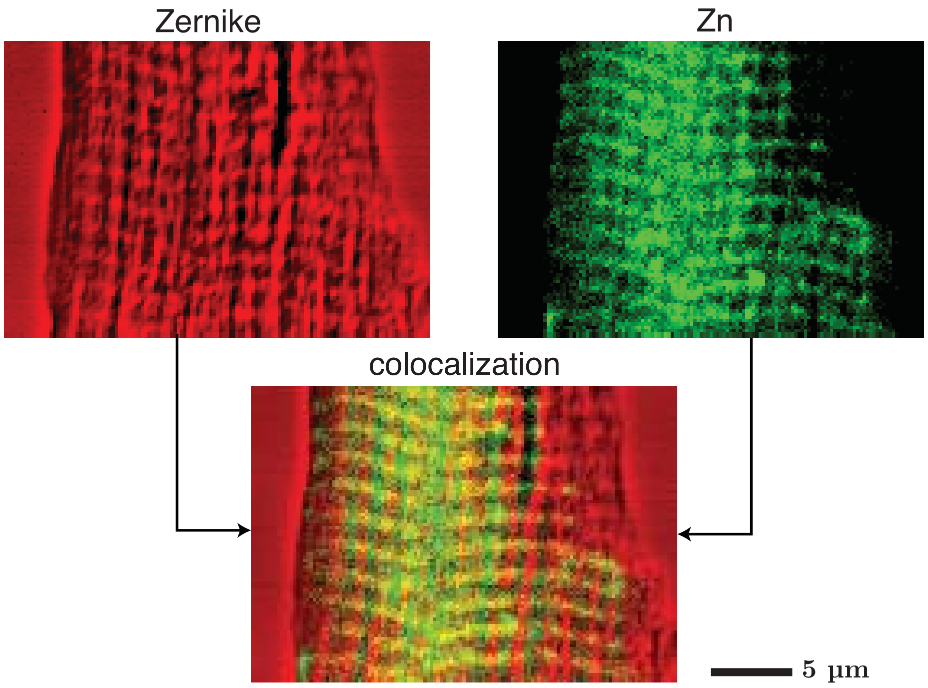




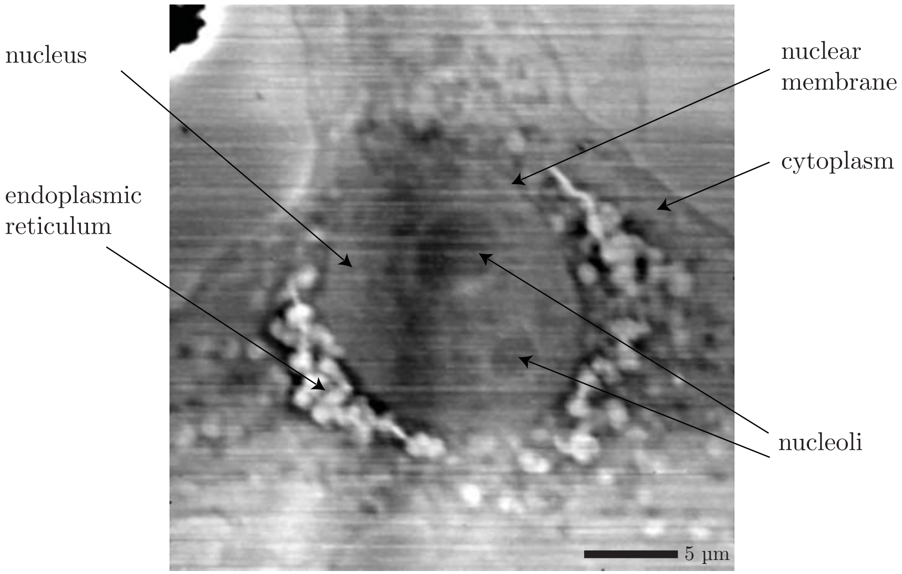
C. Holzner, M. Feser, B. Hornberger, S. Vogt, C. Jacobsen, *Nature Physics* (online: doi:10.1038/nphys1765)

Examples of phase contrast in microprobes

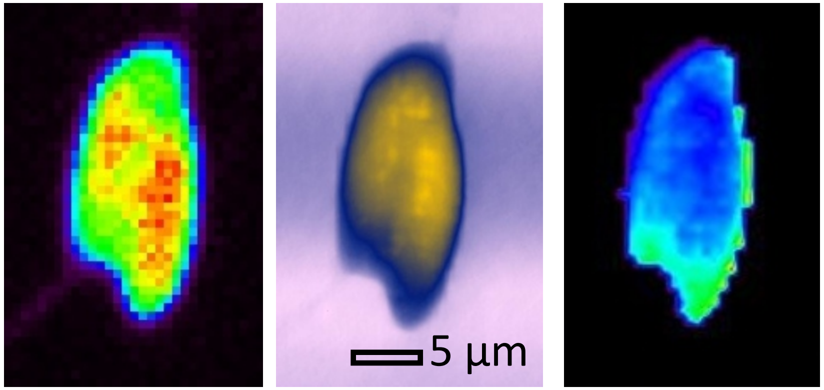
Zinc in Z bands in muscle:
with B. Palmer, U. Vermont



3T3 fibroblast at 10 keV

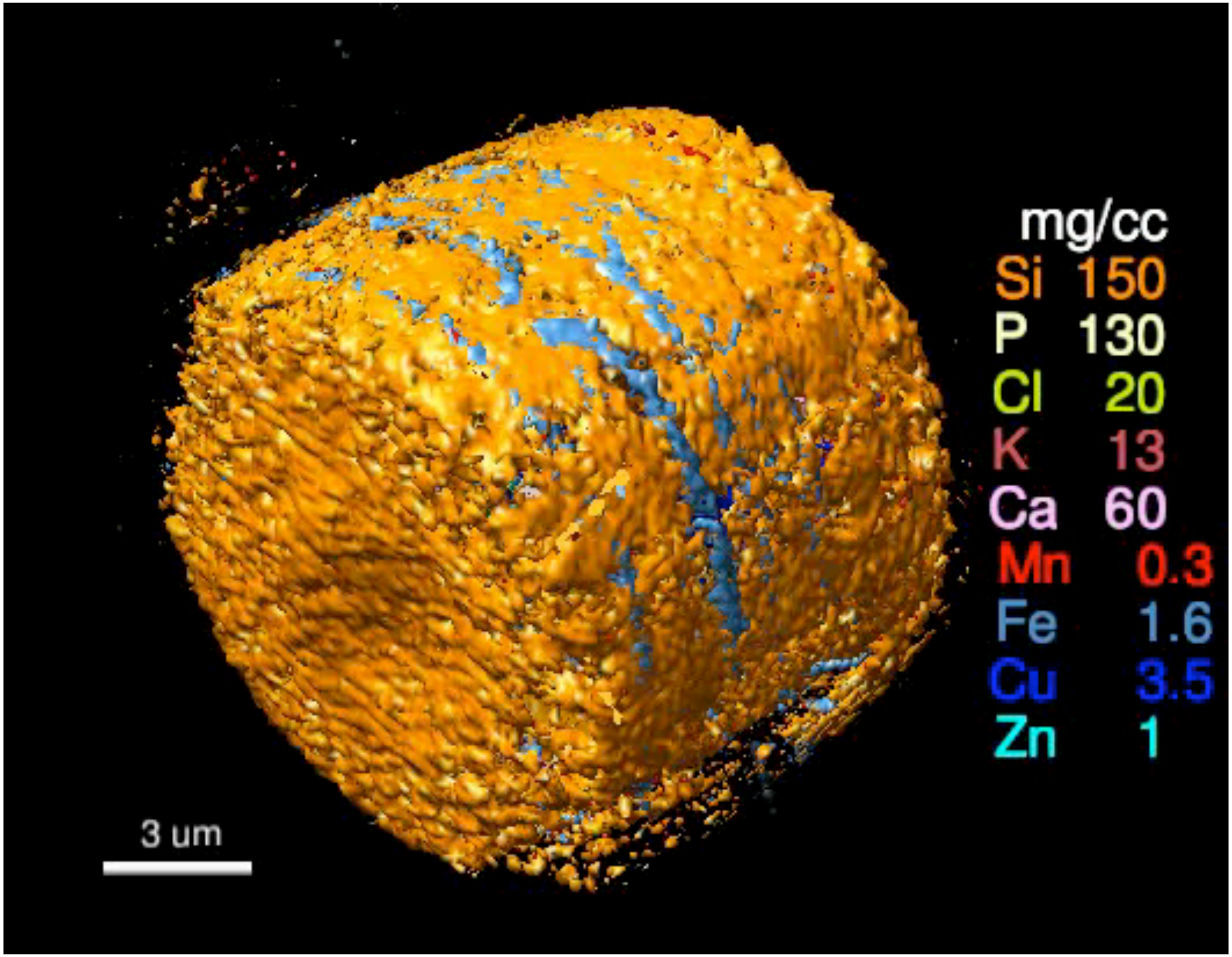


Phase contrast tomography of Cyclotella (marine protist)



Sulfur content Protist mass Sulfur concentration

Fluorescence tomography



de Jonge *et al.*, *Proc. Nat. Acad. Sci.* **107**, 15676 (2010). Next: phase contrast for alignment, dose fractionation for fluorescence.

Christian Holzner on working with Pavel

PhD, August 2010: “Hard x-ray phase contrast microscopy - techniques and applications”

Pavel’s impact beyond

- Segmented detectors are becoming popular!
  - NSLS X1A (Sue Wirick, through August 2010)
  - Three beamlines at the Advanced Photon Source, Argonne
  - Microprobe beamline at the Australian Synchrotron in Melbourne (now operating)
  - Nanoscopium beamline at Soleil in Orsay, France (under construction)
- Enabling lots of good science!

More importantly: impact on people!

- Contributions to our group's intellectual environment, and impact of NSLS X1A (Sue Wirick)
- Three award-winning PhD students!
  - Werner Meyer-Ilse award (best student work at international x-ray microscopy conference): Feser 2002, Holzner 2010.
  - Julian David Baumert award (best PhD work at NSLS): Hornberger 2007.
- Xradia Inc.: commercial developer of x-ray microscope systems, with sales of about $15M/year (half exports?).
  - Michael Feser (Vice President for Advanced Development)
  - Benjamin Hornberger (Product Manager, Synchrotron Systems)
  - Christian Holzner (Staff Scientist, R&D)

Pavel on technology

Pavel: bureaucratic rules, and personal rules

Thank you, Pavel!

Remembering Pavel

Pavel – We remember you!

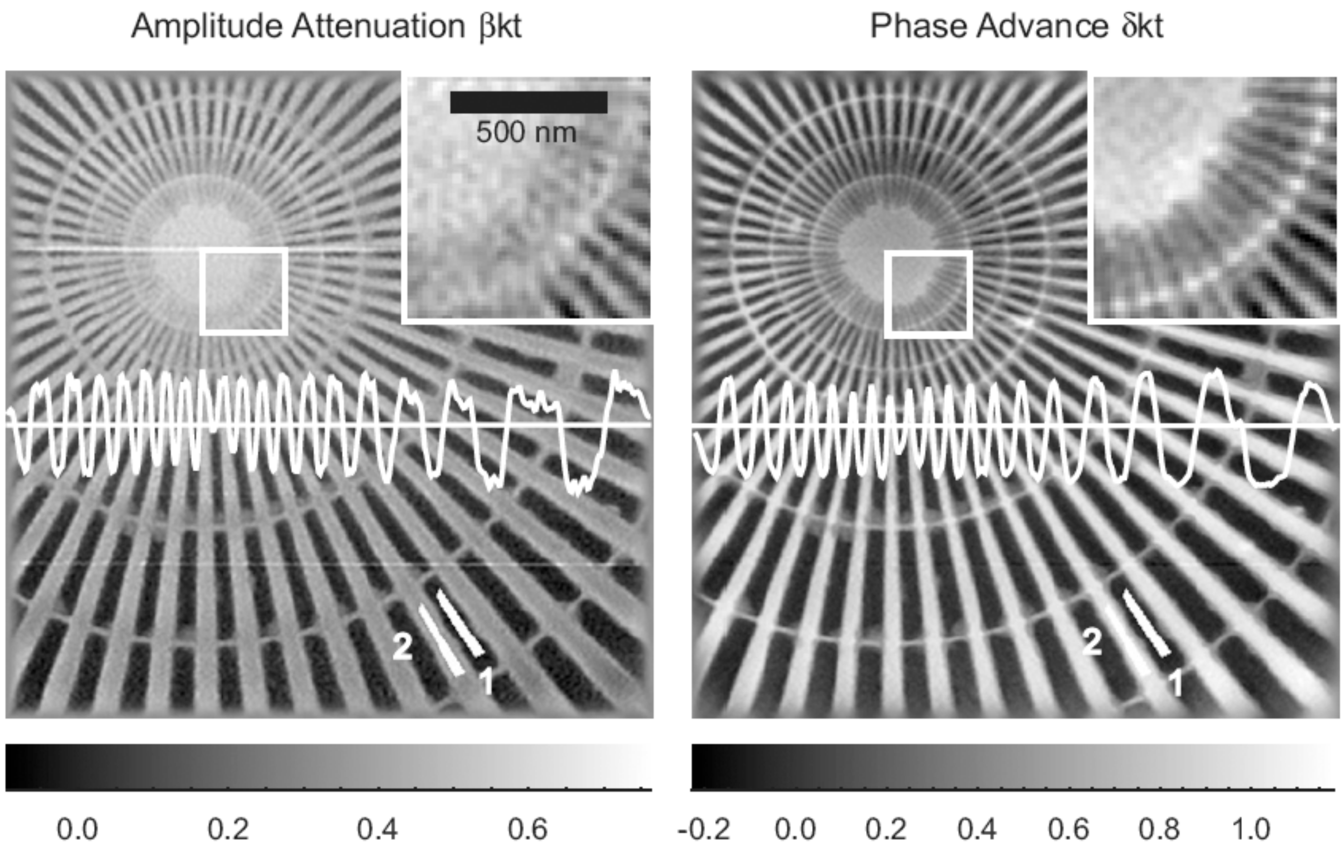
- A brilliant physicist
- An extraordinary personal teacher and mentor
- Long hours taking measurements in the clean room
- Infinite patience for slow learners
- But lots of anger about computers and bureaucrats
- Swimming a mile every day, come what may
- Proficient in seven languages
- Lots of seminar questions

It was a great pleasure working with you!

Michael, Christian and Ben, plus the Stony Brook team

Quantitative phase reconstruction

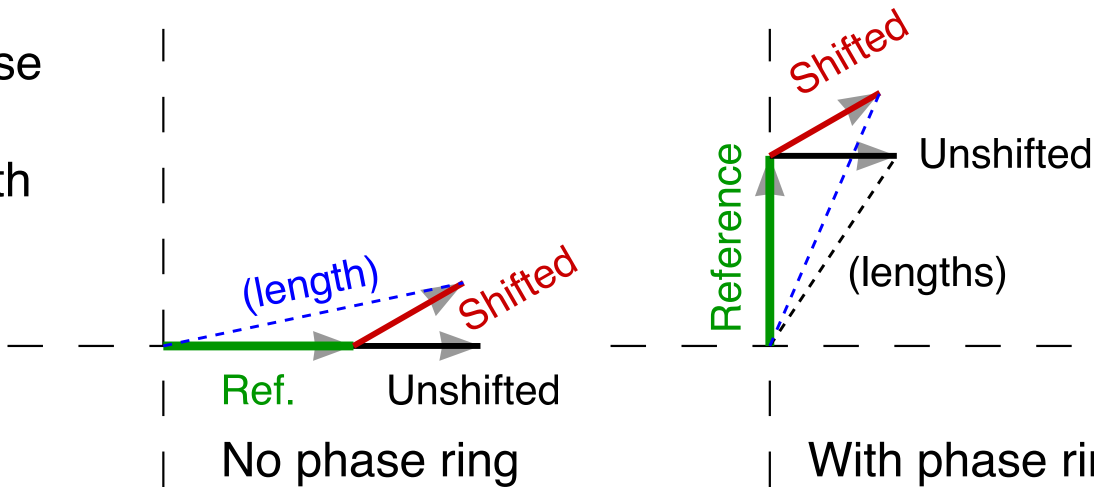
- Hornberger, Feser, and Jacobsen, *Ultramic.* **107**, 644 (2007).
- Fourier filter applied to segmented detector data at 525 eV.
- Quantitative agreement with Henke data.

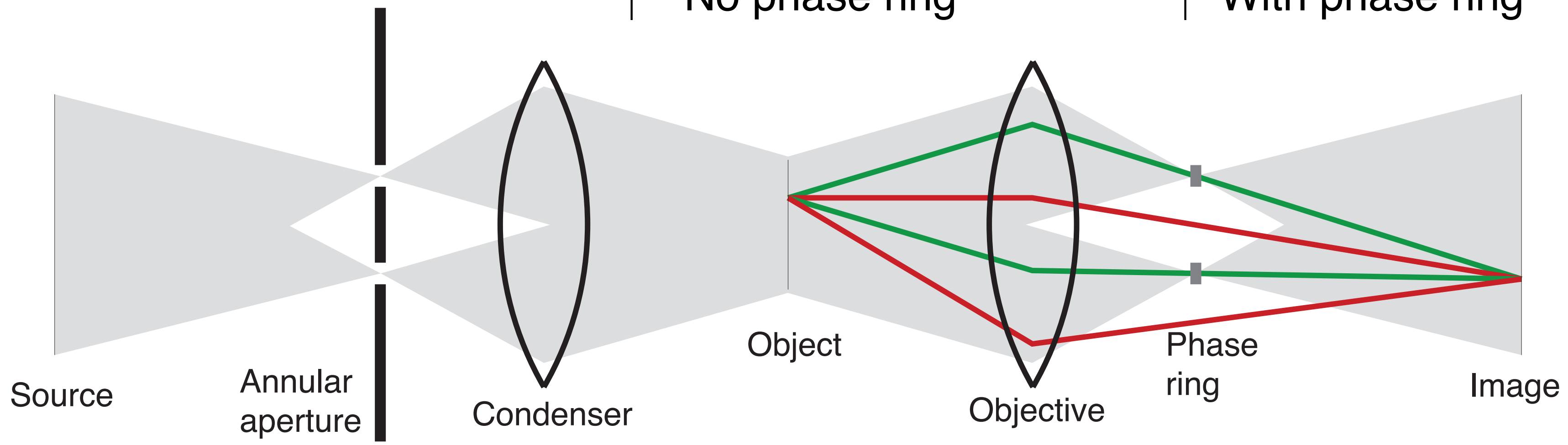


Zernike phase contrast

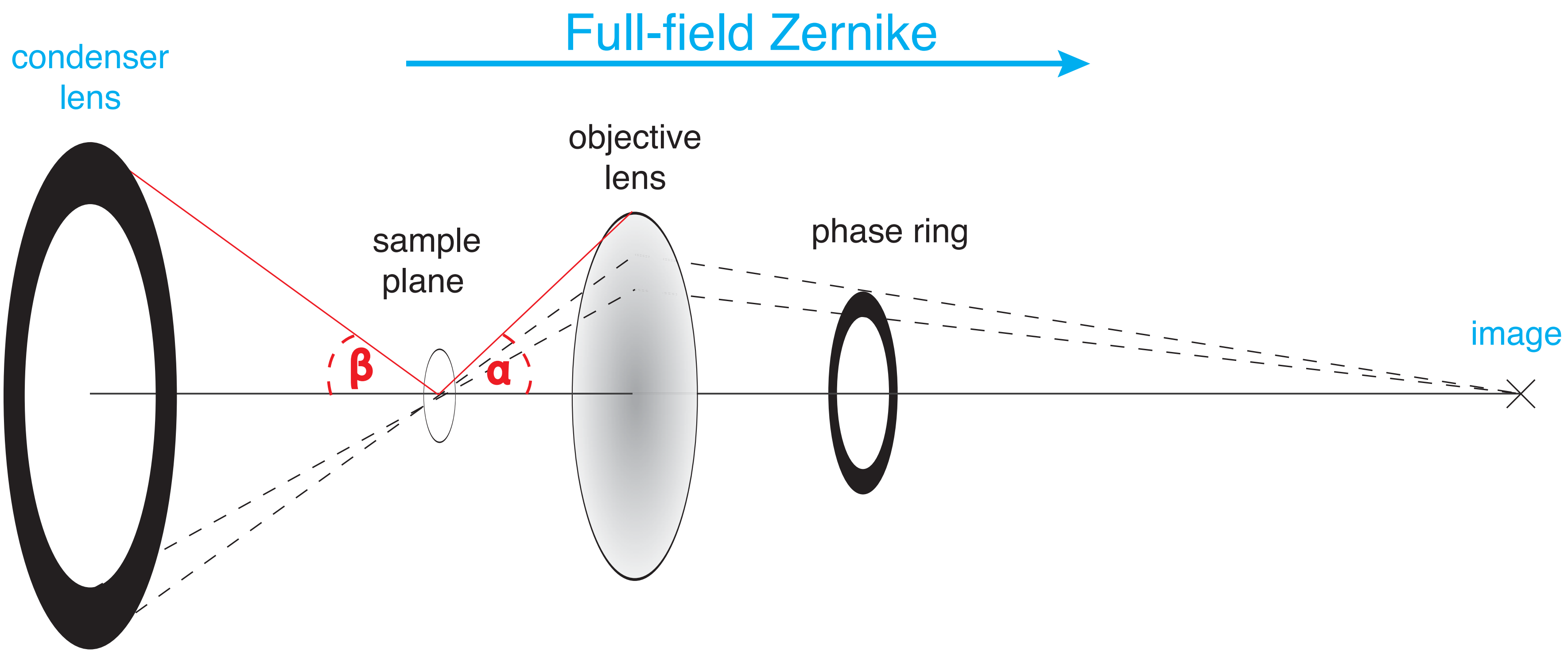
- You don't need a fully coherent source for phase contrast!
- As many coherent "modes" as pixels in detector

Change phase into intensity by mixing with a reference wave



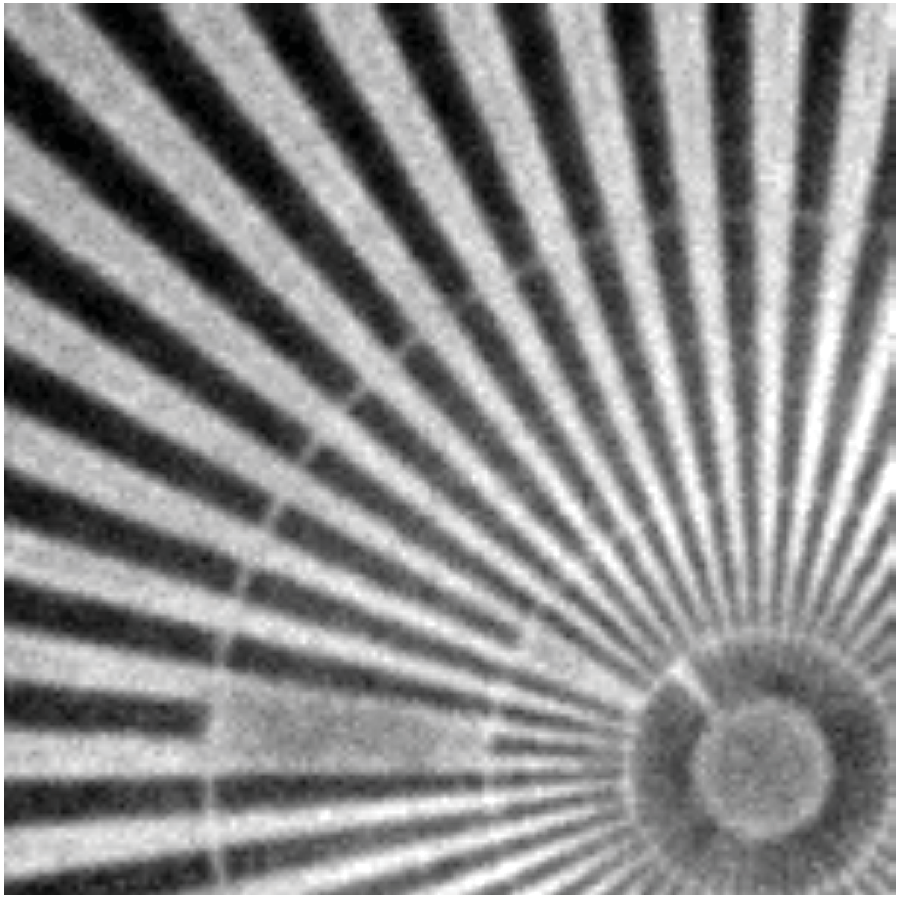


Zernike in a full field microscope



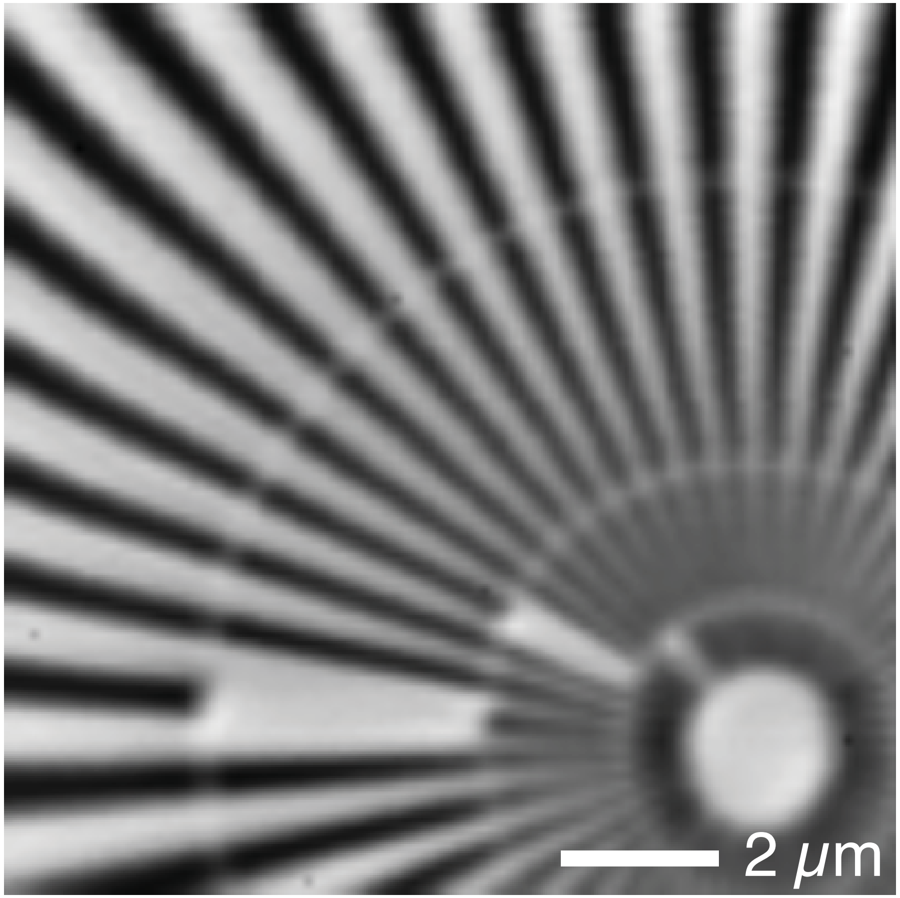
Scanning Zernike phase contrast

full-field



Xradia nanoXCT lab tool at 8.04 keV

scanning



APS 2-ID-E at 10 keV

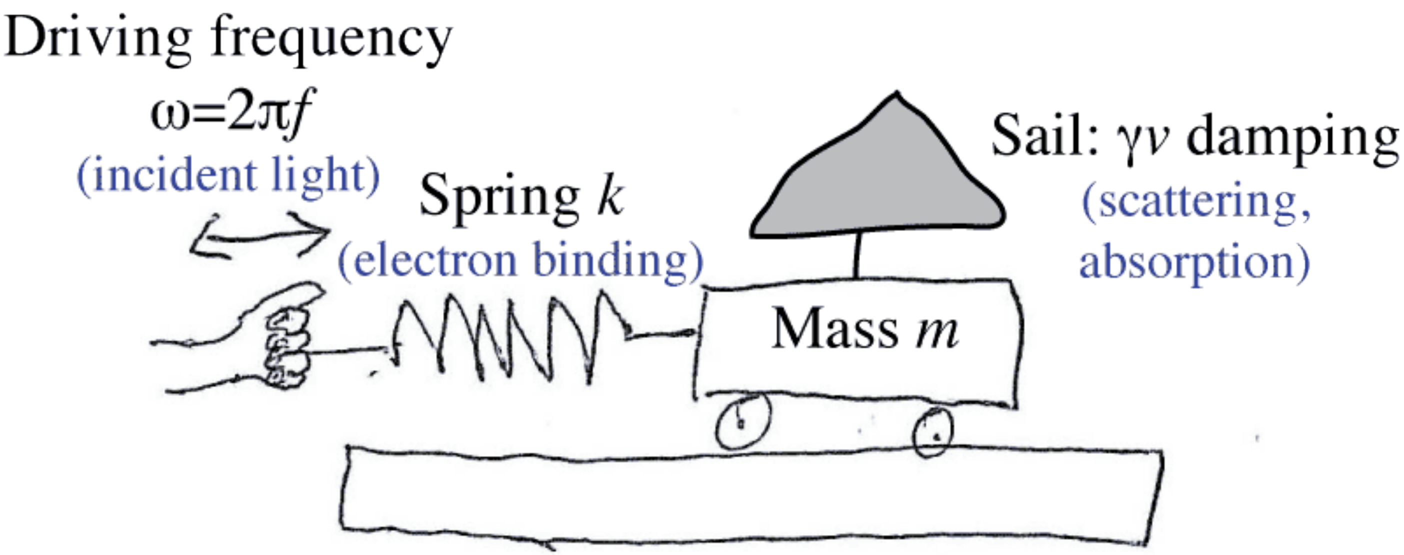
C. Holzner, M. Feser, B. Hornberger, S. Vogt, C. Jacobsen, *Nature Physics* (online: doi:10.1038/nphys1765)

Background to meeting Pavel: x-ray microscopy at the National Synchrotron Light Source

- Soft x-ray scanning transmission x-ray microscopes (STXMs) at the the NSLS.
  - Team included Janos Kirz (Stony Brook: til ~2001), Harvey Rarback (Brookhaven: 1985-1990), Malcolm Howells (Brookhaven: til ~1986), Chris Buckley (Stony Brook, 1988-1990), Sue Wirick (Stony Brook: from ~1995), myself (1988-2009), and lots of postdocs and students.
- U-15 (~1982-1987): bending magnet beamline
  - 300 nm/1 eV/1 second per pixel.
- X-17t (1986-1987): 10 period undulator
  - 70 nm/0.5 eV/0.2 seconds per pixel.
- X-1A (1989-2010): 37 period undulator
  - eventually 30 nm/0.1 eV/0.005 seconds per pixel.

Some background to X-ray phase contrast

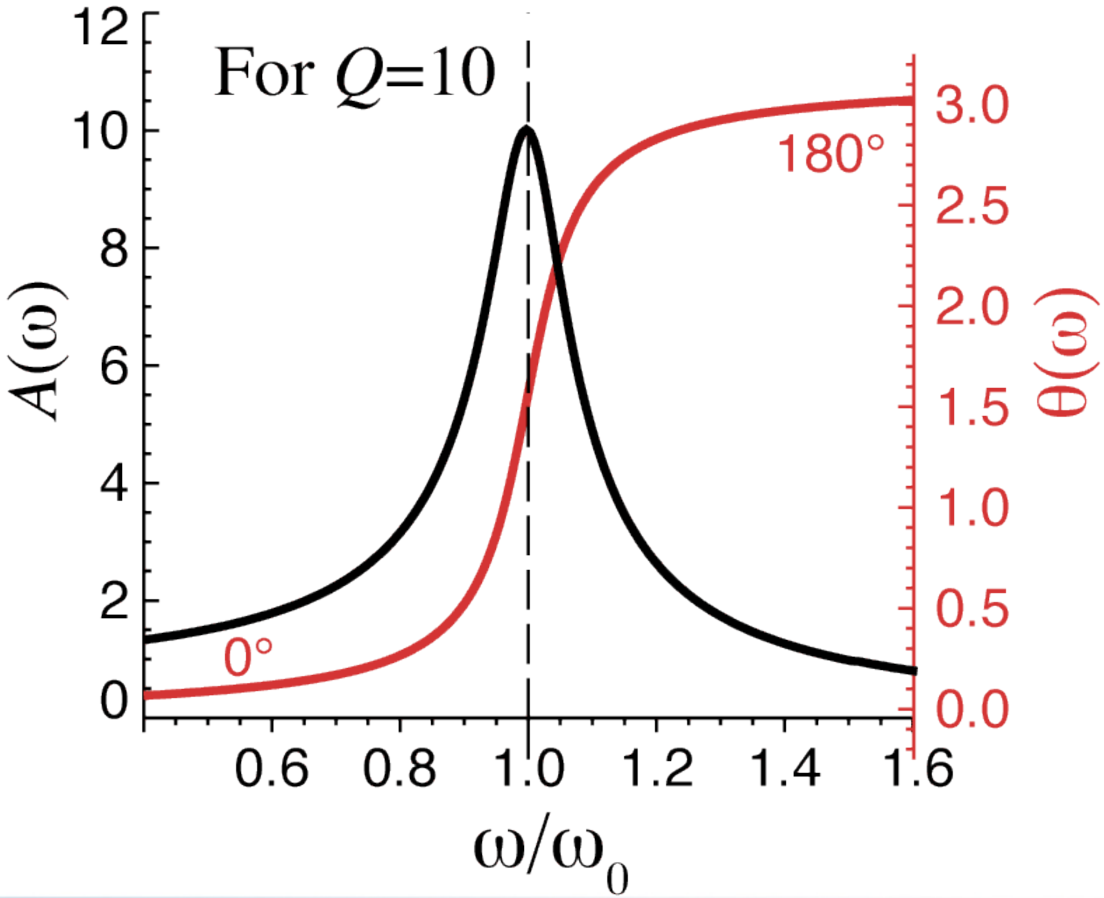
- Damped, driven harmonic oscillator



- Damping: scattering, absorption
- Driving: incident electromagnetic wave ω
- Harmonic oscillator: electron quantum state with energy $\hbar\omega_0 = \hbar\sqrt{k/m}$

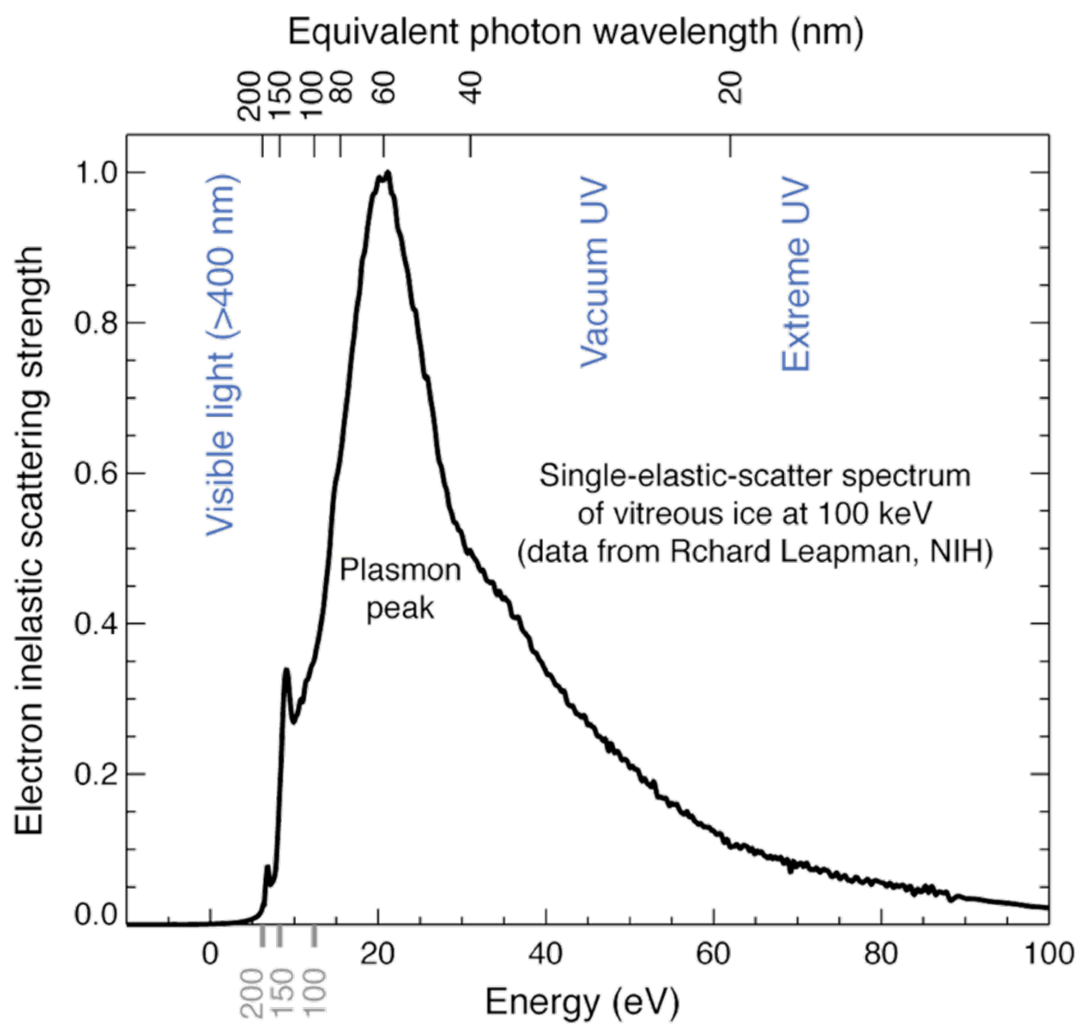
Damped, driven harmonic oscillator

- Single resonance: absorption peak, phase shift across resonance
- Q=resonant frequency/damping=ω_0/γ

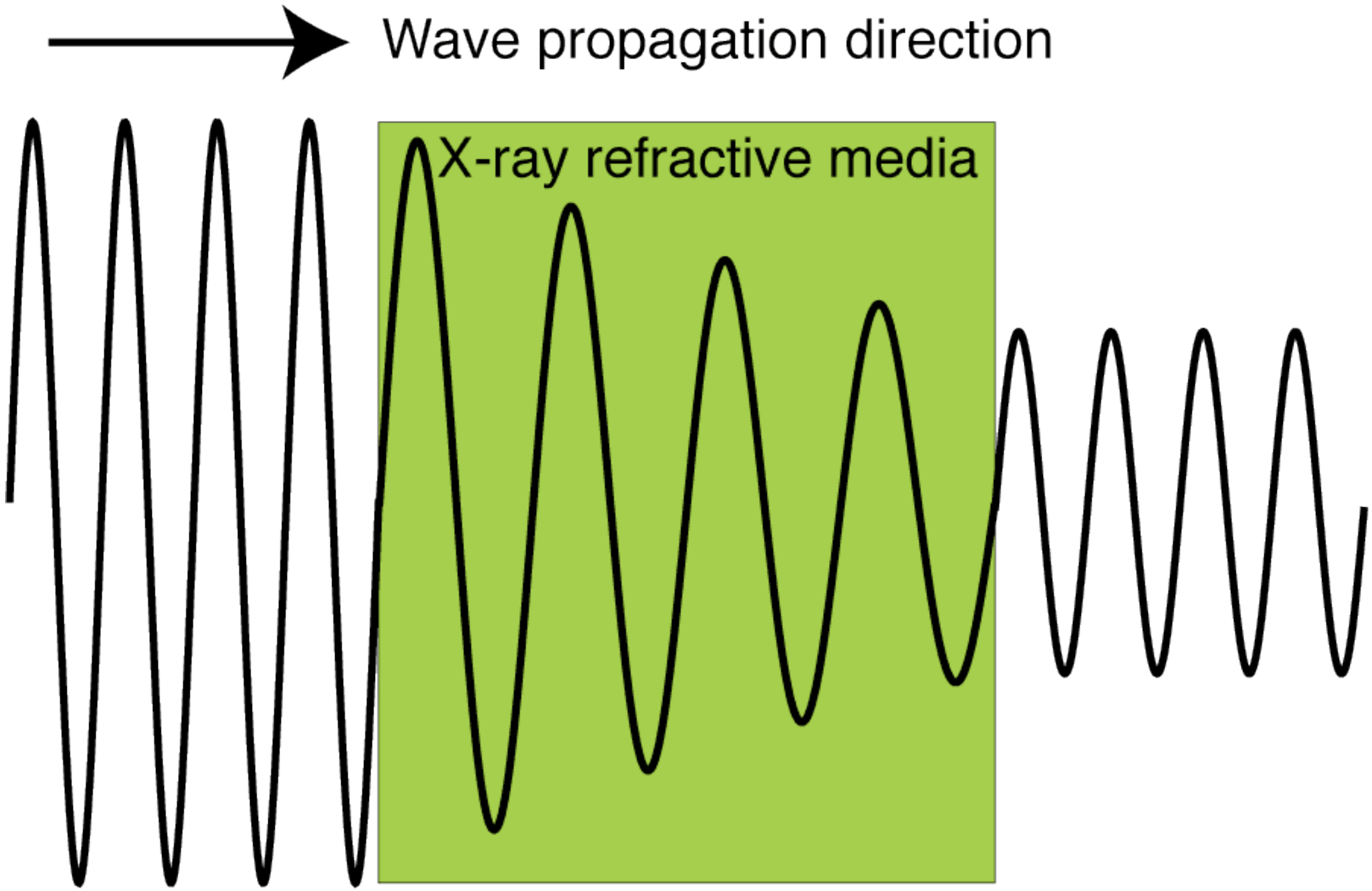


The preponderance of plasmons

Dividing line between low and high frequency limits of refractive index



X rays in media



(Benjamin Hornberger)

Mysteries of the refractive index

Write refractive index as

$$n = 1 - \frac{n_a r_e}{2\pi}\lambda^2\left(f_1 + if_2\right) = 1 - \alpha\lambda^2\left(f_1 + if_2\right)$$

Phase velocity is faster than light in vacuum!

$$v_p = \frac{w}{k} \approx c\left(1 + \alpha\, f_1\lambda^2\right)$$

Prisms refract x rays the opposite way from visible light!

Phase is advanced rather than retarded!

Total external reflection with critical angle

$$\theta_c \approx \sqrt{2\alpha\,\lambda^2 f_1}$$

But group velocity is OK:

$$v_g = \frac{dw}{dk} \approx c\left(1 - \alpha\, f_1\lambda^2\right)$$

86 A. Einstein, [Nr. 9/12.

Lassen sich Brechungsexponenten der Körper für Röntgenstrahlen experimentell ermitteln?

Von A. Einstein.

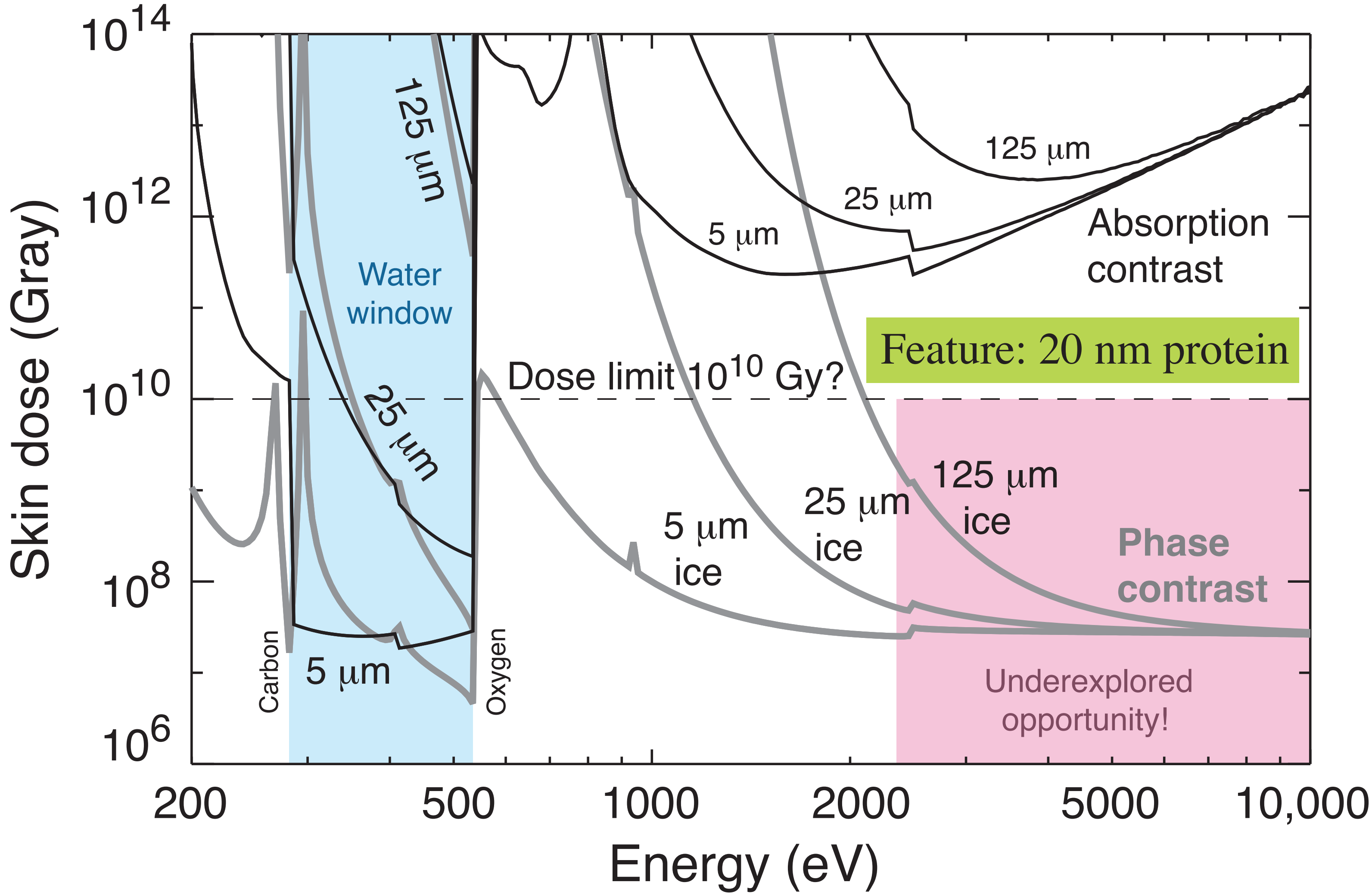
(Eingegangen am 21. März 1918.)

Vor einigen Tagen erhielt ich von Herrn Prof. A. Köhler (Wiesbaden) eine kurze Arbeit[1]), in welcher eine auffallende Erscheinung bei Röntgenaufnahmen geschildert ist, die sich bisher nicht hat deuten lassen. Die reproduzierten Aufnahmen — zumeist menschliche Gliedmaßen darstellend — zeigen an der Kontur einen hellen Saum von etwa 1 mm Breite, in welchem die Platte heller bestrahlt zu sein scheint als in der (nicht beschatteten) Umgebung des Röntgenbildes.

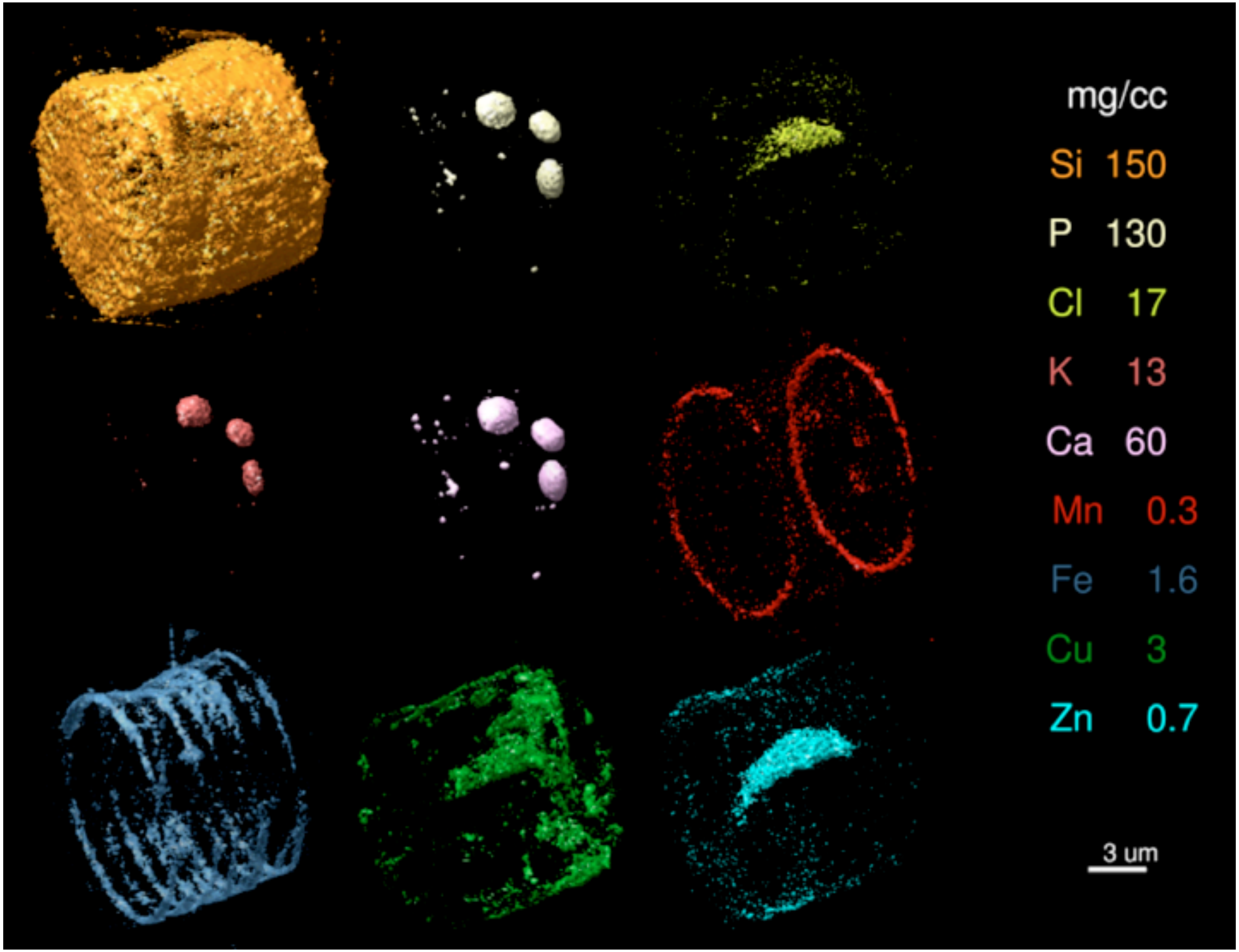
Ich möchte die Fachgenossen auf diese Erscheinung hinweisen und beifügen, daß die Erscheinung wahrscheinlich auf Totalreflexion beruht. Nach der klassischen Dispersionstheorie müssen wir erwarten, daß der Brechungsexponent n für Röntgenstrahlen nahe an 1 liegt, aber im allgemeinen doch von 1 verschieden ist. n wird kleiner bzw. größer als 1 sein, je nachdem der Einfluß derjenigen Elektronen auf die Dispersion überwiegt, deren Eigenfrequenz kleiner oder größer ist als die Frequenz der Röntgenstrahlen. Die Schwierigkeit einer Bestimmung von n liegt darin, daß $(n - 1)$ sehr klein ist (etwa 10^{-6}). Es ist aber leicht einzusehen, daß bei nahezu streifender Inzidenz der Röntgenstrahlen im Falle $n < 1$ eine nachweisbare Totalreflexion auftreten muß.

Radiation dose for 100% efficient imaging

Phase contrast is good in the "water window", and essential at higher energies



Quantitative 3D fluorescence of a diatom



M. de Jonge, C. Holzner, S. Baines, B. Twining, K. Ignatyev, J. Diaz, D. Howard, A. Miceli, I. McNulty, C. Jacobsen, S. Vogt, *Proc. Nat. Acad. Sci.* **107**, 15676 (2010)